Molecular Conductors

The Critical Magnetic Field of the Superconductor λ-$(BETS)_2GaCl_4$ with the Field Parallel to the Conducting Planes

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We have measured the penetration depth of λ-$(BETS)_2GaCl_4$ (BETS) as a function of field with the applied field parallel to the conducting planes. The sample was placed inside a small coil that was connected in parallel to a capacitor, and made to self resonate at 45 MHz with a tunnel diode. The magnetic field of the small coil was perpendicular to the conducting planes of the BETS. We relate the rf penetration to the frequency of the self resonant circuit using a simple formula, and find the superconducting transition using a method similar to the standard methods used to find the superconducting transition with resistance measurements. The H_{C2} derived from either measurement yields the same value.

We have preliminary H_{C2} data for the BETS salt with the applied magnetic field parallel to the conducting planes that shows $H_{C2//}(0) \approx 14$ T in contrast to the $H_{C2\perp}(0) = 2.9$ T. The H-T phase diagram shows there is a positive slope change, or kink in the parallel data at the same temperature as in previously measured perpendicular data. This new data suggests that the kink is not dependent on magnetic field direction and is temperature induced. The upturn in the phase line occurs at ~T/Tc = 0.4. Four possible explanations exist for this type of phase diagram. They are dimensional crossover, strong coupling, a magnetic transition, or a change in the superconducting order parameter.

We have ruled out the first three phenomena for various reasons, some based on the fact that the upturn or kink in the phase diagram exists at all orientations. Thus, we are now considering the explanation of the kink in the phase diagram as a change in the superconducting order parameter. A similar phase diagram was seen in the UPt_3 heavy Fermion superconductor. It was determined that the symmetry of the order parameter changed in UPt_3 as a function of temperature. The evidence for this type of transition in BETS is not yet convincing, however, qualitatively the change in slope of the H-T phase line is consistent with a change in the order parameter. Consider a case where the symmetry of the order parameter changed from s-wave to d-wave. The s-wave portion for a BCS superconductor starts as a straight line, and develops a slightly negative curvature as the temperature decreases. D-wave superconductors, at least the HTSC layered materials that are somewhat similar to the 2D organics, have a strong positive curvature. Therefore, it is consistent to have a change in the slope of the H-T phase line if the superconductor changes the symmetry of its order parameter.

Magnetoresistance in $(DMET\text{-}TSeF)_2X$ System

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The title material is a novel material developed from the similar basis of two other charge-transfer salts TMTSF and DMET salts. X are anions ($X=AuCl_2$, AuI_2,…), which contrary to Bachgaard salts, are all linear, and do not introduce an anion ordering.[1] At ambient pressure, both mentioned materials exhibit metallic behavior of resistivity, but while superconductivity (SC) is found in AuI_2 sample, in the $AuCl_2$ sample SC is not confirmed down to 50 mK. Both materials, however, show a series of magnetic transitions characteristic for field induced spin density wave (FISDW) obtained in related Bachgaard salts. We have performed first electrical transport measurements in fields up to 30 T and temperatures down to 50 mK. Typical magnetoresistance data for $AuCl_2$ sample at 0.6 K, for a set of angles (magnetic field to c-axis) is shown in Figure 1. The kinks in resistance curves are typical for FISDW transitions observed previously in Bachgaard salts.[2] The field for the onset of these FISDW transitions is much higher than in $(TMTSF)_2ClO_4$ material (only member of Bachgaard salts superconducting at ambient pressure), but the "frequency" of 53 T, and its angular (orbital) dependence (Figure 2) reveal the same origin of these transitions in both materials.

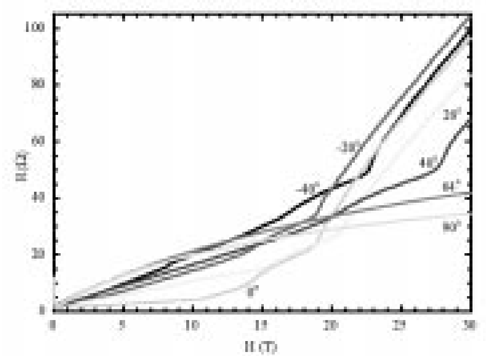

Figure 1. Resistance [R] vs. magnetic field (H) of $(DMET\text{-}TseF)_2AuCl_2$ for a set of angles.

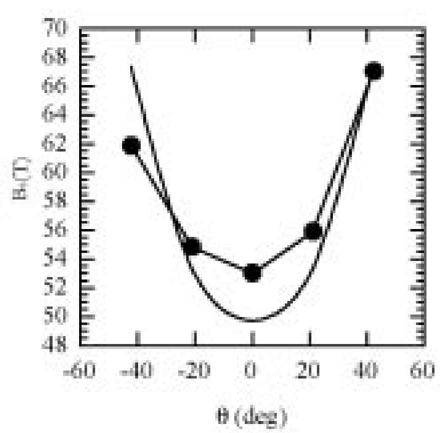

Figure 2. Frequency of FISDW transitions (B_S) in tesla as a function of angle (θ).

The detailed (angular, temperature, magnetic and electric field) measurements are currently in progress.

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High Field Magnetization of the Spin-Peierls Compound $(TMTTF)_2PF_6$

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$(TMTTF)_2PF_6$ is a half-filled quasi-one dimensional organic material in which electronic correlations open up a relatively small charge gap, Δ_ρ~600K.[1] The spin excitations remain ungapped until T < 17 K, below which a spin-Peierls (SP) state is established.[2] Above a critical field B_c, the dimerized SP state becomes unstable to localized spin excitations because of the Zeeman energy. The spin excitations are S=1/2 (on a single chain) but are created in pairs. The spin excitations are also associated with a π phase shift of the lattice distortion, so that the high field phase is incommensurate with wavevector determined by the applied field.

In recent ^{13}C NMR experiments,[3] the onset of the high-field phase of $(TMTTF)_2PF_6$ was found to be approximately 19 T. The signature was a broadened NMR spectrum, signaling that the spin excitations were also pinned to the lattice.[3]

We carried out high-field cantilever magnetization measurements to determine the spin density distribution characteristic of the high field phase.[4] A summary of the main results appears in Figure 1. Each curve is a field sweep, constructed by subtracting the data obtained from a sweep at center-field position and then dividing by the magnetic field itself to obtain results proportional to the magnetization. The top curve is a field sweep at a temperature above T_{SP}. It shows nearly no net force (or magnetization) up to fields greater than 24 T, from which we conclude that the paramagnetic and Larmor diamagnetic contributions are nearly equal in the high temperature phase. The observed curvature arises from a systematic error in subtracting the center-field sweep from the out-of-center result. The next trace, taken well below T_{SP} at 7 K, still contains the diamagnetic contribution. At low fields the paramagnetic contribution no longer exists or is very small. At about 19 T, the paramagnetic contribution starts to increase, indicative of the critical field B_c. As for other SP systems such as $CuGeO_3$, $M=\chi B$ at high fields, where χ is the constant spin susceptibility of the uniform phase. That is, only the low field magnetization is altered by the presence of the spin gap. The highest curve is offset from the others and exhibits a hysteresis loop that is more than 6 T wide at this temperature. There was no significant hysteresis for T > 4 K.

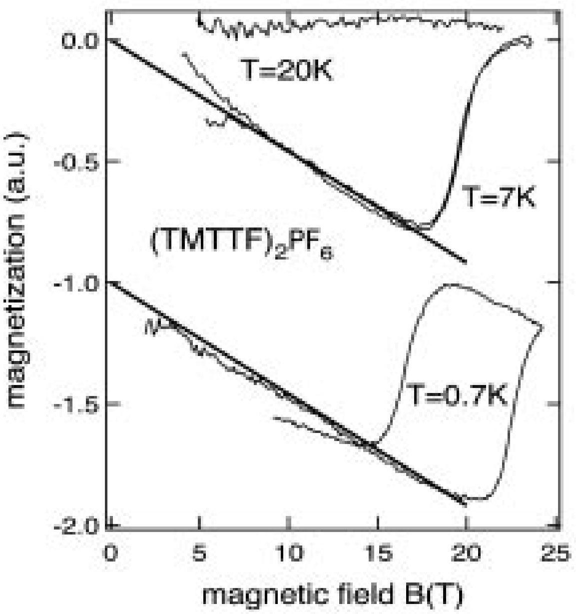

Figure 1. Magnetization M vs. applied field B for representative temperatures. The data recorded at T=0.7 K is offset from the others.

This work was supported in part by NSF under grants DMR-9705369 (WGC) and CHE-9257081 (CAM).

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Incommensurate Phase of the Organic Spin-Peierls Compound $(TMTTF)_2PF_6$

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The Spin-Peierls (SP) ground state is a non-magnetic dimerized phase forming in some quasi-1D antiferromagnetic insulators. At high magnetic fields, the singlet SP phase is unstable because of the Zeeman energy, and in one dimension, magnetic excitations in the form of soliton/anti-soliton pairs are expected to be spontaneously created.[1] The solitons have S=1/2 and are actually anti-phase domain walls when referenced to the phase of the lattice distortion.

The known SP systems exhibit very similar magnetic field/temperature phase diagrams. At high temperatures is the paramagnetic uniform (U) phase. Below T_{SP} the onset of a continuous drop of the magnetic susceptibility signals the transition to the dimerized (D) Spin-Peierls phase. Above a critical field $B_c \approx k_B T_{SP}/g_{\mu B}$, the creation of localized magnetic excitations with repulsive interactions results in an incommensurate (I) phase.

Here we report a ^{13}C NMR study at low temperatures and fields near to B_c for $(TMTTF)_2PF_6$. The

TMTTF molecules were ^{13}C spin-labelled on the sites bridging the dimer.[2]

Figure 1 illustrates the effect on the ^{13}C lineshape from increasing the field. At the top, the applied field is somewhat below B_c, and the line is narrow because of the small χ_s. Small increases just above B_c lead to broadening along with the gradual diminishment of the narrower central feature.

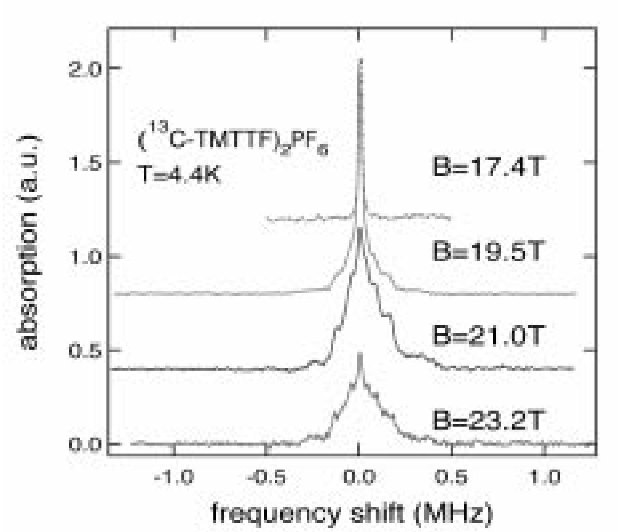

Figure 1. ^{13}C spectra collected at different fields B near to the critical field B_c.

With one important exception, the results are not very different than predictions.[3] Here, the contributions from the two unique ^{13}C sites make the assignment of a local field distribution difficult. In particular, we identify in all of the spectra several "ledges" on each side of the central feature. We are unable to account for all of these features using the simplest model where the lattice distortion vanishes at the center of the domain wall and each chain is identical. We are able to clearly identify exactly four features on the positive side and three or four on the negative side of the central feature. The existence of four ledges might indicate an unusual period doubling of the spin density which persists through the domain wall.

This work was supported in part by the NSF under grants DMR 9705369 and CHE-9257081.

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High Field NMR Studies of Conduction Electron Dynamics in Metallic Polypyrrole-PF_6

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This work probes the electron spin dynamics of metallic polypyrrole-PF6 doped to 25% PF6 using $1/T_1$ of protons as a test of whether the electron transport is metallic on a microscopic scale. Measurements were made at 0.96 GHz (temperature T = 1.7-55 K) and 0.38 GHz (1.7-300 K). Since the effects of molecular motion are expected to be of negligible importance,[1] the main mechanisms expected to produce $1/T_1$ in doped polypyrrole are translational motion of the conduction electrons and purely orientational motion of localized electron spins, which contribute via nuclear spin diffusion to these paramagnetic centers (SDPC).[2] An important advantage of our high field measurements is that the SDPC process freezes out below about 15 K, where the local moment magnetization saturates.

Our more highly doped sample (s1) is a free-standing film prepared using the procedures of Yoon *et al.*[3] Its dc electrical conductivity varies smoothly from 100 S/cm at 1.5 K to 220 S/cm at 280 K. In addition, a less highly doped sample (s2) with a lower electrical conductivity was investigated at 0.38 GHz.

Figure 1 shows ($1/T_1T$) as a function of T. The vertical arrows indicate where the electron Zeeman energy is $2k_BT$, below which the SDPC process becomes frozen out. Although the observed behavior indicates that it may contribute up to about 50% of the total rate, substantially more work is needed to establish what fraction of the total rate is caused by it. The mechanism for the observed decrease in $1/T_1T$ above 150 K has not yet been identified; it is, however, consistent with more rapid electrical transport at high temperatures. The low T increase in $1/T_1T$ for the less highly conducting sample (s2) probably reflects a relatively longer correlation time for electron motion in that sample at low temperatures.

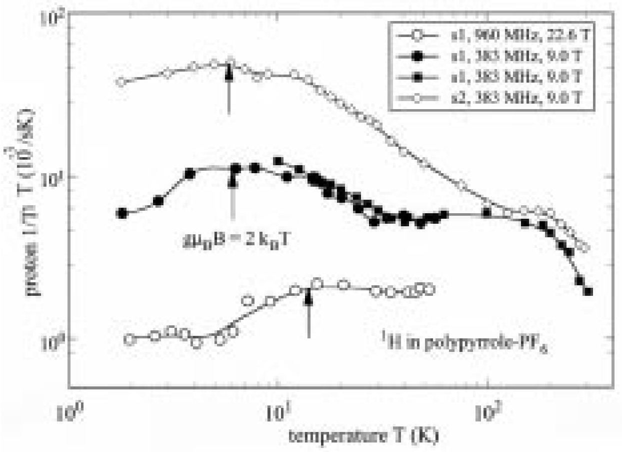

Figure 1. Proton $1/T_1T$ as a function of T for high density metallic polypyrrole-PF6. The solid lines are a guide to the eye.

Our most important result is that a substantial contribution to ($1/T_1$) persists to the lowest temperatures, where the only viable mechanism for it is translational motion of the conduction electrons. Since this contribution appears to contribute significantly at all temperatures as expected of a metallic state, we conclude that the electrical transport in this material is metallic on a microscopic scale. On the other hand, the observed frequency dependence is not the power law 0.5 expected of 1d diffusive electron motion on an infinite chain.

The part of the work done by the UCLA participants was supported by NSF Grant DMR-9705369 (UCLA Physics) and LANL CULAR Grant 9819 (UCLA Chemistry).

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NMR Investigation of Spin Density Wave Critical Fluctuations in $(TMTSF)_2PF_6$

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One of the unresolved questions regarding the spin density wave (SDW) transition in Bechgaard salts is the appropriate description of the transition and its fluctuations. Here, we report measurements of the proton spin-lattice relaxation rate ($1/T_1$) in the vicinity of the transition temperature (T_N) over an unusually wide range of frequencies. By carrying the measurements to nearly 1 GHz, it is possible to investigate a very broad range of frequencies in the critical fluctuation spectrum. The results discussed here are NMR measurements of the contribution of the spin density wave (SDW) transition critical fluctuations to $1/T_1$ in $(TMTSF)_2PF_6$. They were carried out at 14.9 MHz in an electromagnet at UCLA and 0.98 GHz in a resistive magnet at Tallahassee.

The critical behavior close to T_N is shown in Figure 1, where $1/T_1$ is plotted as a function of $|T\text{-}T_N|^{\theta}$. The solid line, which corresponds to the power law $1/T_1 \propto |T\text{-}T_N|^{-0.75}$, is a best fit to the 0.98 GHz data above T_N. Because no substantial background subtraction is required for this data set, it provides the best resolution in these measurements. Although the 14.9 MHz data above T_N and the 0.98 GHz data below it have a much lower resolution, within the experimental error, they are consistent with the same exponent.

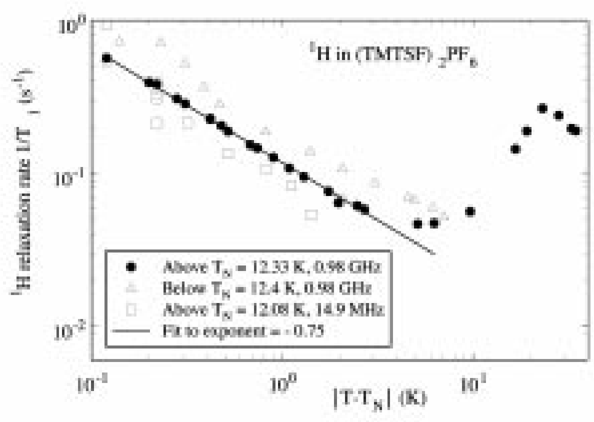

Figure 1. Critical behavior for $1/T_1$ of protons in $(TMTSF)_2PF_6$. The solid line, which corresponds to the exponent -0.75, is the best fit to the 0.98 GHz data above T_N.

The most important observations from these measurements are that in the critical regime, $1/T_1$ is nearly independent of the magnetic field alignment and the frequency on both sides of the transition and that it varies as $|T\text{-}T_N|^{\theta}$ with $\theta = -0.750 \pm 0.08$. This independence of $1/T_1$ on frequency and orientation indicates that the critical fluctuations have a magnetic field orientation that is isotropic and that their correlation time is less than 2×10^{-10}s for all conditions of these measurements.

Our observed power law for $1/T_1$ in the critical regime is consistent with dynamical scaling for the 3D Heisenberg model of Bourbonnais.[1] In this case, $\theta = \gamma - D\nu + z\nu$, where γ is the critical index for the staggered magnetic susceptibility, ν is the critical index, and z is the dynamical exponent. From the values ν = 0.60–0.63, γ = 1.25–1.30, and z = 2.0 + a small correction,[2] one obtains θ in the range 0.62–0.71, which has some overlap with our measured value θ = 0.75±0.08. On the basis of this result, we conclude that our measurements of $1/T_1$ in the critical regime are consistent with dynamic scaling for the 3D Heisenberg model on both sides of the transition. This behavior is very different from that observed in the ordered SDW phase, where the dominant coupling is to SDW phasons whose characteristic times cover a very broad range.[3,4]

The part of this work done by the UCLA participants was supported by NSF Grant DMR-9705369.

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Thermodynamic Observation of Magnetic-Field Induced First-Order Phase Transition in α-$(BEDT\text{-}TTF)_2KHg(SCN)_4$

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In experiments at the NHMFL in 1998, we have been investigating the magnetic-field dependence of the low temperature ground states in the quasi-2D molecular conductor α-$(BEDT\text{-}TTF)_2KHg(SCN)_4$, and related materials using two complementary thermal measurements: the field dependent specific heat and the magnetothermal effect. Because of the quasi-2D nature of the material, we have also been investigating the dependence of both measurements on the angle of the magnetic field relative to the conducting planes. These measurements have been done in a miniature rotatable calorimeter designed to fit into two of the dilution refrigerators at NHMFL: (1) the top-loading dilution refrigerator with a 20 T superconducting magnet (and rotating probe), and (2) the top-loading "portable" dilution refrigerator that fits into the 33 T resistive magnets. The initial development and testing of the calorimeter was done while NAF was visiting at NRIM.

In both the specific heat and magnetothermal measurements, we measured the amplitude of a sinusoidally varying sample temperature while slowly varying the applied magnetic field. For the specific heat measurements, we modulated the sample heater power output; for the magnetothermal measurements, we instead quickly modulated a small additional applied magnetic field.

In both α-(BEDT-TTF)$_2$KHg(SCN)$_4$ and κ-(BEDT-TTF)$_2$Cu(NCS)$_2$, we have observed magnetoquantum oscillations in both the specific heat and magnetothermal effect with fundamental frequencies (and harmonics) in good agreement with previously known transport measurements.

In α-(BEDT-TTF)$_2$KHg(SCN)$_4$, however, we also found two pieces of thermodynamic evidence for a phase transition above 23 T at 0.35 K, where a kink occurs in the magnetoresistance. In the specific heat,[1] we find that the non-oscillatory background contribution is field-independent between 23.4 T and 33 T, consistent with a transition into a high field metallic state. In our magnetothermal measurements,[1] we also find magnetocaloric evidence for a phase transition at 23.4 T, indicating that this high field transition is first order. Additional small hysteretic anomalies of unknown origin are observed between 26 T and 33 T. A dramatic change in oscillation amplitude in both the specific heat and the magnetothermal effect occurs above 23 T at 0.35 K, consistent with previous SdH[2] and dHvA[3] measurements. Note that there is no direct signature of a phase transition in the dHvA data at this field, although there is a clear increase in the effective mass.

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Role of Electron-Electron Interactions in Organic Conductors

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In connection with numerous current attempts to find experimental proof for a non-Fermi liquid behavior in quasi-one-dimensional conductors of the Bechgaard Salts family, we undertook an effort to interpret the available data for the temperature dependence of resistivity in one of these materials (TMTSF)$_2$PF$_6$. The material undergoes the low temperature transition into an insulating spin density wave (SDW)-state. By applying pressure (about 6 to 7 kbar), however, this transition may be suppressed and the material remains in the normal state down to ~1K.

The data exist from a few different groups and are quite reproducible. At the first glance, there are considerable deviations from the T^2-behavior expected in the Fermi-liquid theory.

It was shown that this "non-Fermi liquid" feature may be easily explained if the "nesting" structure of the electronic spectrum of these materials is taken into account. Thus, the conclusion is that strong electron-electron interaction is not necessary, at least, for understanding of the anomalous temperature dependence of resistivity in (TMTSF)$_2$PF$_6$.

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Gor'kov, L.P., *et al.*, Phys. Rev. B, 57, 6204 (1998).

Ensemble Dependence of the de Haas-van Alphen Effect in α-$(ET)_2KHg(SCN)_4$

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We investigated the magnetic breakdown effect (MB) in a quasi-2-dimensional organic conductor, α-$(ET)_2KHg(SCN)_4$, with a full quantum mechanical tight binding model. In previous reports,[1] we provided a natural description of combination frequencies, especially the clear appearance of the semiclassically forbidden frequency ($\beta-\alpha$) in the de Haas-van Alphen effect (dHvA). There have been several reports[2-4] that the forbidden frequency is a result of the chemical potential oscillation in canonical ensemble. The purpose of this report is to verify that combination frequencies and the forbidden frequency have a quantum mechanical origin, and appear in *both* canonical and grand canonical ensembles.

In our ensemble dependent calculation, thermodynamic quantities were calculated using the field dependent energy spectrum. The thermodynamic potential Ω is given by

$$\Omega = -\frac{2}{\beta}\sum_{\vec{k}}\sum_{\alpha=1}^{4}\sum_{\gamma=1}^{\tilde{q}} \ln[1+e^{-\beta(E_\alpha^\gamma(\vec{k})-\mu)}] \qquad ,$$

where α is the band index, γ is the magnetic subband index, and β is $1/(k_BT)$. In the canonical ensemble, the number of carriers is fixed and the chemical potential is determined from the conservation of the total number of electrons. Since there are 6 electrons per unit cell and 4 electronic bands, the occupancy factor N/N_{max} is 3/4.

With the thermodynamic potential and the chemical potential calculated, the free energy, $F = \mu N+\Omega$ was obtained.

The magnetization with fixed number of particles (M_N), and with fixed chemical potential (M_μ,) were calculated by differentiating M_N=-dF/dB and M_μ=-dΩ/dB, respectively. The magnetization in both cases is a similar saw-tooth shape with inversed fields (Figure 1). In Figure 2, the Fourier transformed spectrum of dHvA oscillations is plotted. Both cases show the same frequencies with variations in the amplitudes. The fundamental frequency (f_α) and the MB frequency (f_β) are 610 T and 4230 T, respectively. In both cases, combination frequencies, such as $f_\beta + f_\alpha$ and $f_\beta - f_\alpha$ are observed with several harmonics of f_α and f_β.

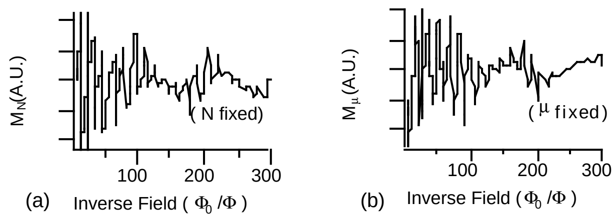

Figure 1. The magnetization as a function of inverse magnetic field. (a) M_N is the magnetization with fixed *N* (b) M_μ is the magnetization with fixed μ.

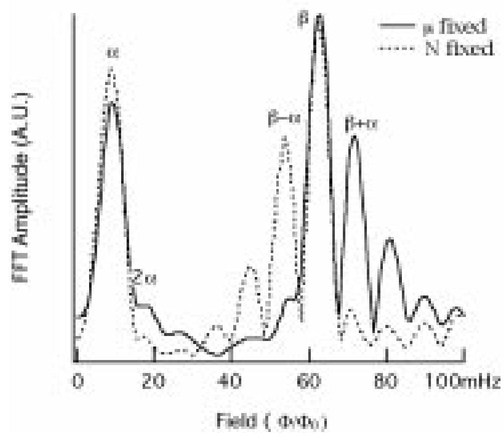

Figure 2. The ensemble dependent FFT result. The dotted line and the solid line indicate the canonical and the grand canonical ensembles, respectively.

In conclusion, the ensemble dependent calculation of MB in our model has demonstrated the existence of forbidden frequencies in *both* canonical and grand canonical ensembles. This result verifies the origin of the combination orbits, including forbidden orbits, as the mixing between the magnetic subbands associated with the two partially occupied bands.

This work was supported by NSF-DMR 95-10427 (JSB).

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Uniaxial Stress Studies of the SDW State in $(TMTSF)_2PF_6$

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We report uniaxial stress/strain studies on $(TMTSF)_2PF_6$, which at ambient pressure and temperatures below 12 K enters a spin density wave (SDW) ground state. Previous hydrostatic pressure experiments[1-4] have shown that the nesting conditions are suppressed with applied pressure. Theoretical approaches, using a mean field approximation,[5] predicts an anisotropic dependence of hopping parameters on the SDW gap (Δ). Therefore, a uniaxial stress experiment is a very useful tool to probe this anisotropy.[6] Stress has been applied along all three of the principal axes *a*, *b*, and *c*.

The temperature dependent electrical resistance was monitored in the temperature range of 30 K to 4.2 K. It shows a metallic behavior in the higher temperature region, and at ambient pressure the typical 12 K density wave transition. The SDW transition temperature (T_{SDW}), and the activation energy (Δ) were obtained from Arrhenius plots at each pressure along the three different crystal axes. Both T_{SDW} and Δ decreased with increasing stress.

We note that for all 3 axes, the reductions in the resistance under uniaxial pressure were as drastic as the case of hydrostatic pressure experiments, although the changes in T_{SDW} diminished, we have not yet been able to stabilize the metallic state, which we estimate to occur around 20 kbar.

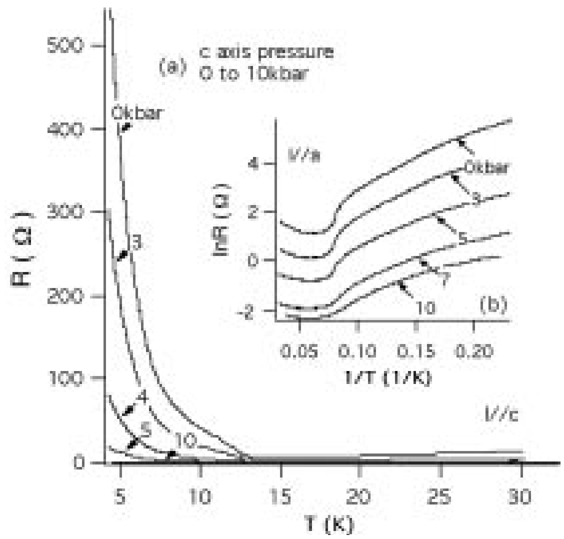

Figure 1. c axis stress, 0 to 10 kbar for I//c and I//a (a). R vs. T for I//c, (b). lnR vs. Inverse T for I//a.

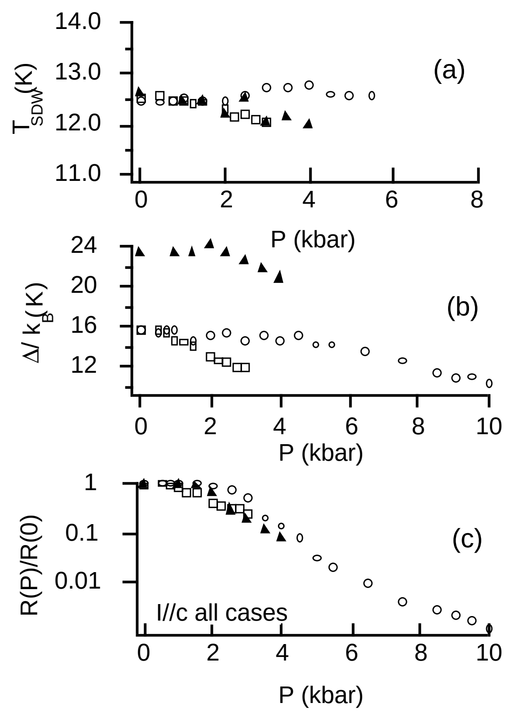

Figure 2. (a) T_{SDW} vs. pressure. (b) Reduction in resistance vs. pressure at 4.5 K (z; a axis, c; b axis, µ; c axis). In all cases the current is parallel to c direction.

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Frequency Mixing of the Magnetic Breakdown Oscillations and Their Temperature-Dependence in the Canonical Ensemble

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Magnetic breakdown is a phenomenon that has been known about for many years,[1] occurring in materials with two or more different Fermi surface sheets separated by small gaps. Historically, this has been studied in three dimensional metals with multiply connected Fermi surfaces, and the subject was essentially brought to a close with the publication of a paper by Falicov and Stachowiak some years ago.[2] Their elegant theory was able to explain the magnetic field-dependence of all the combination frequencies that are observed in such materials.

This all changed when low-dimensional organic charge-transfer salts entered the scene, for several reasons: their low-dimensionality tends to imply that the oscillations in such quantities as the density of states, transport scattering rate, and quasi-classical mean free path, are much larger than in normal metals. This implies that novel magnetotransport effects, such as Stark quantum interference, are very readily observed.[3] This also implies that the oscillations of the chemical potential are much larger than in normal metals. For several years, the presence of so-called "anomalous" frequencies, namely β-α and β-2α, in the magnetotransport and magnetization, have been the subject of controversy.[3] It is difficult, however, to make any firm conclusions on the basis of magnetotransport data alone. Only when one compares magnetotransport data with magnetization data can one see that there are certain quantum oscillation frequencies prominent only in the magnetotransport that can, therefore, only be attributed to magnetotransport-related mechanisms (rather than thermodynamic mechanisms).[3] More often than not, these are due to Stark quantum interference. This is pretty much well established now.[3, 4]

The more surprising fact was the presence of "anomalous" (or forbidden) frequencies, albeit of weak amplitude, in the magnetization.[5] This was surprising because the magnetization is after all a thermodynamic function of state, and one should, therefore, not expect frequencies that are forbidden by the Falicov-Stachowiak theory. Numerical models were then, however, able to show that such forbidden frequencies arose quite naturally as a result of frequency mixing effects due to the oscillations of the chemical potential.[3] An elegant analytical expression for this effect was soon developed by Fortin and Ziman, yet was limited in that it was valid only at zero temperature and in infinitely pure samples (i.e. with no Landau level broadening).[6] The theory, therefore, overestimated the amplitudes of the various frequencies observed in real samples.

By applying the numerical methods[3] to a wider range of temperatures, it becomes very clear in Figure 1 that the "forbidden" combination frequencies β-α and β-2α are radically damped at higher temperatures compared to the parent frequencies α and β, giving the appearance of a heavy mass. The rapid suppression of these frequencies at finite temperatures, and with quasi-particle scattering, explains why these frequencies are observed to be very weak in the magnetization.[5]

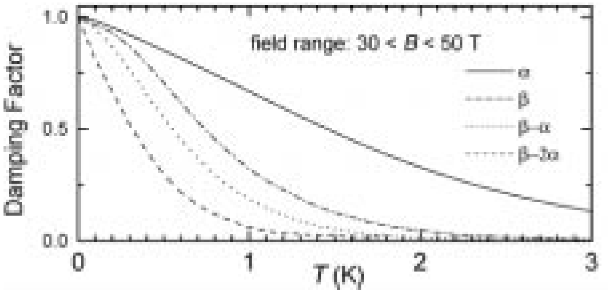

Figure 1. The temperature dependencies of the normal parent frequencies β and α (allowed by the Falicov-Stachowiak theory), and the mixed frequencies β-α and β-2α that come about due to the oscillations of the chemical potential in the canonical ensemble.

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The Fermi Surface of α-$(BEDT\text{-}TTF)_2KHg(SCN)_4$

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Of central importance to the physical understanding of the density wave formation, and the quantum Hall effect in α-BEDT-TTF$)_2$KHg(SCN)$_4$ (and related salts), is a detailed knowledge of the Fermi surface topology. A large body of angular magnetoresistance measurement data and quantum oscillation data now exists, clearly demonstrating that the topology of the Fermi surface departs somewhat from that originally calculated by Mori, *et al.*[1,2]

In order to obtain an improved model for the Fermi surface and band structure, we developed a method, by which, the parameters of the tight-binding Hamiltonian can be fitted to the experimentally determined Fermi surface. The resulting fit that best represents the experimentally determined Fermi surface details is shown in Figure 1. Note the small gap between the one-dimensional sheets and the two-dimensional pocket (the α pocket), which is necessary in order to explain the observed magnetic breakdown.

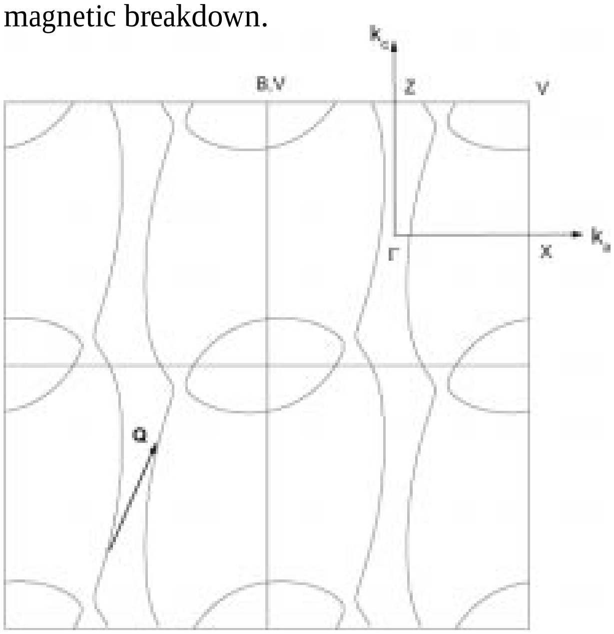

Figure 1. The calculated Fermi surface after fitting the Hamiltonian to the experimental Fermi surface data.

From this fitted Fermi surface, we can deduce only one commensurate nesting vector of the form $Q=k_a/5+xk_b+2k_c/5$ that is capable of nesting part of the strongly warped one-dimensional sheets. The reconstructed Fermi surface that results from such a nesting vector is shown in Figure 2. It turns out that this form of the reconstructed Fermi surface is then able to explain all of the de Haas-van Alphen and quantum interference frequencies that are observed experimentally.[3] In particular, one of the quantum interference frequencies corresponds to an area in k-space equal to the reconstructed Brillouin zone. The new one-dimensional sheets that are formed due to the Fermi surface reconstruction account for the one-dimensional angular magnetoresistance oscillations that are observed in the density wave phase.

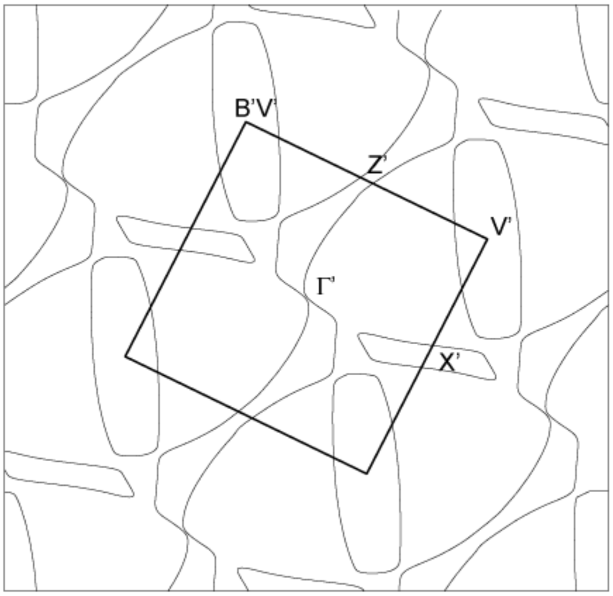

Figure 2. The reconstructed Fermi surface (and the new Brillouin zone) after the application of the periodic potential, as described in the text.

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The Phase Diagram of α-Phase Charge-Transfer Salts

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α-phase charge-transfer salts, such as α-(BEDT-TTF)$_2$KHg(SCN)$_4$, have been studied for the last decade, and the debate still continues as to whether the low temperature, low magnetic field ground state, characterized by a reconstructed Fermi surface topology, constitutes a spin density wave (SDW) or a charge density wave (CDW). This is partly due to the fact that the necessary diffraction experiments have not been performed in order to distinguish these two possibilities.

All of the earlier publications stated quite firmly that these materials possessed an SDW. Evidence for antiferromagnetism appeared to come from measurements of the anisotropic susceptibility at low temperatures.[1] However, this could simply reflect the fact that the Landau diamagnetic component of the magnetism is anisotropic, and that this degree of anisotropy changes upon reconstruction of the Fermi surface. The only other evidence for an SDW would then be the μSR results. However the claimed spontaneous moment of 3x10^{-3} μ_B is at the limit of the resolution of the experimental technique, and is much smaller than that observed in TMTSF salts.[2] According to the theory of SDWs, this would then require an effective Coulomb repulsion that is much larger than the total electronic bandwidth of the material. When we also consider the fact that NMR measurements have provided no evidence for any substantial internal magnetic field, the arguments in favor of an SDW ground state begin to look very tenuous.[3] On top of all this, the low temperature, low magnetic field ground state is destroyed by a magnetic field at ~ 23 T (i.e. at the “kink” transition field). Such a behavior is incompatible with the theory of SDWs in a magnetic field.[4] The destruction of the low temperature, low magnetic field ground state in a magnetic field is nevertheless exactly what one should expect for a CDW, since a CDW is composed of singlet electron-hole pairs.[5] The kink transition field is also about the same order as one expects according to Clogston’s argument, and is also observed to be isotropic.

Considering the ground state of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ to be driven by CDW rather than SDW order, starting with the gap equation in the presence of a magnetic field, we can obtain an expression for the free energy of the system. The CDW nesting can be considered only to take place for the 1D sheet (i.e. the Fermi surface consists of both a 2D pocket and a pair of 1D sheets), and the 2D pocket can be assumed to remain relatively unaffected. Although the two Fermi surface sheets are virtually independent, they are still thermodynamically coupled by the chemical potential μ. The oscillations of μ driven by the changing Landau quantization of the 2D pocket in a magnetic field, can then have dramatic

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consequences on the gap equation and the free energy. At zero temperature, Clogston's original argument is modified by μ so that

$$h = \sqrt{\frac{\Delta^2}{2} - \tilde{\mu}^2}$$

where $h=\mu_B gB/2$ is the reduced critical field, Δ is the order parameter and $\tilde{\mu}$ is the oscillatory component of μ. Because $\tilde{\mu}>\Delta$, instead of there being only a single transition, the oscillations of μ cause there to be a cascade of first order transitions repeating at a rate that is twice the frequency of the 2D α pocket, $2F_\alpha$. This could then explain the pronounced hysteresis that is observed. The overall phase diagram expected by this model is given in Figure 1. A $\tilde{\mu}$ term continues to contribute to the free energy of the CDW phase, at all fields below the first order transitions. This can then explain the pronounced 2nd harmonic of the α frequency that is observed within the low temperature, low magnetic field phase. Instead of it originating with spin-splitting of the Landau levels, it originates from oscillations in the Pauli paramagnetic susceptibility of the nested 1D sheets. Indeed, experimentally, many features of the oscillations, including the over-pronounced 2nd harmonic, had already been shown to be inconsistent with the spin-splitting model.[6]

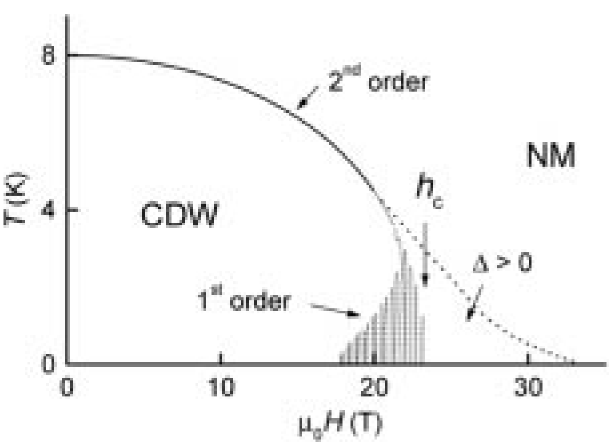

Figure 1. The phase diagram of α-(BEDT-TTF)$_2$KHg(SCN)$_4$, according to the Pauli-limited CDW model, affected by oscillations of μ coming from the α pocket. At high temperatures, the transition is of 2nd order, while at high fields the transition(s) are of 1st order.

Figure 2 shows the total free energy of the 1D and 2D sheets over a large range of magnetic field at zero temperature. Note the appearance of a strong 2nd harmonic of the α frequency oscillations within the low temperature, low magnetic field phase without needing to invoke any spin-splitting effects.

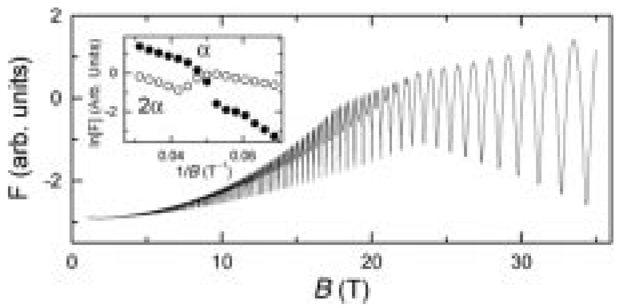

Figure 2. The calculated free energy of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ as a function of magnetic field. Note that the oscillations are split within the CDW phase, although this has nothing to do with spin-splitting.

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High Field Electrodynamic Investigation of $(TMTSF)_2ClO_4$

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A cavity perturbation technique was used to study the phase diagram of $(TMTSF)_2ClO_4$ at low temperatures and high magnetic fields. Details of the experimental technique are described elsewhere.[1] The sample was oriented within a cylindrical cavity with its *c** axis parallel to the applied DC magnetic field, and currents were excited in the *ab* plane of the sample. Figure 1 shows the dissipative and reactive response of the sample at ~44 GHz.

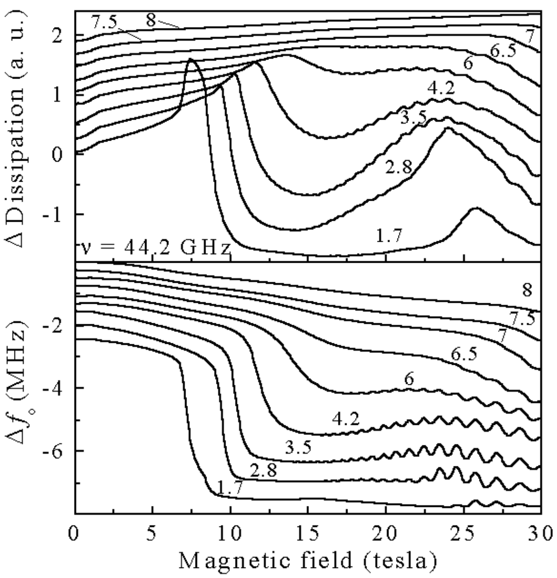

Figure 1. Electrodynamic response as a function of temperature.

This behavior may be understood as follows. At low fields, the monotonic increase in the dissipation simply reflects the magneto-resistance of the sample. The weak inflection at low fields (1 T to 3 T) is due to a novel form of quasi-one-dimensional cyclotron resonance (see Ref. 2). At the lowest temperatures, the field region from about 5 T to 8 T corresponds to all but the last of the well known field-induced-spin-density-wave (FISDW) phases.[3] The sample is still conducting in this field region. The sharp increase in dissipation seen in the lowest temperature trace is due to the loss of carriers to the FISDW. Careful inspection of this data reveals more than one FISDW transition. Once the final FISDW phase is entered, the sample becomes insulating,[4] and the cavity resonance condition changes significantly—the resonance frequency shifts strongly downwards, indicating that the effective volume of the cavity increases, at the same time, dissipation within the cavity decreases. These facts suggest that the electromagnetic fields within the cavity penetrate the entire sample, and that there is little or no dissipation. This confirms that the radiation frequency (44.2 GHz) is well below the single particle gap and well above the collective mode response of the FISDW. In contrast, when the sample is conducting, dissipation is dominated by the surface resistance.

Although the sample is supposedly insulating, we still observe a significant dissipative peak at high fields, together with quantum oscillations. The present data suggest that a reorganization of the FISDW condensate occurs in the vicinity of the phase boundary at ~28 T.[5] Consequently, we see a significant contribution to the conductivity due to the FISDW, at these frequencies.

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Angle-Dependent Transport Measurements in the Quantum Hall Regime of α-(BEDT-TTF)$_2$TlHg(SCN)$_4$

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The quantum Hall effect (QHE) has been shown to occur in the layered organic conductors α-(BEDT-TTF)$_2M$Hg(SCN)$_4$, M = K, Tl, by the observation of plateaus in ρ_{xy},[1] pronounced eddy current resonances in the magnetization[2, 3] and the saturation of the Hall voltage in pulsed magnetic fields.[4] A phenomenon intrinsically linked with multi-layered quantum Hall systems is the presence of a chiral Fermi liquid (CFL) around the edges of the sample. It consists of a sheath of highly conducting surface states facilitating ballistic transport perpendicular to the layers. The CFL affects the interplane magnetoresistivity whenever the chemical potential lies between adjacent Landau levels, bypassing the conventional current paths through the bulk of the sample. This leads to a suppression of the Shubnikov-de Haas maxima with falling temperature and eventually to a phase reversal of the oscillations in very high quality samples.[3, 4]

The degradation of the QHE at fields above ~ 40 T, observed in α-(BEDT-TTF)$_2$TlHg(SCN)$_4$,[4] has been explained by the onset of magnetic breakdown between the quasi-one- and quasi-two-dimensional sections of the Fermi surface.[4, 5] Since magnetic breakdown will also alter the arrangement of the density of states necessary for the formation of the CFL, a concomitant weakening of the effect of the CFL on the interplane transport is expected at these fields. In Figure 1, we can see that for $\theta = 0^\circ$ and T = 0.8 K, the oscillation phase of ρ_{zz} is inverted immediately upon entering the QHE regime at the kink transition. In this region, the oscillations are in anti-phase to those observed at higher temperatures that can be described by a bulk transport model (*c.f.* trace for $\theta = 0^\circ$ and T = 3 K). The original oscillation phase is recovered after passing through a node at B ~ 38 T. If we identify the strength of the effect of the CFL, with the degree to which the resistance values at fields of integer filling factors are lowered, then the position of the node gives a good indication when the CFL phase is destroyed. A comparison with Hall voltage measurements[4] on the same sample shows that this happens synchronously with the demise of the QHE.

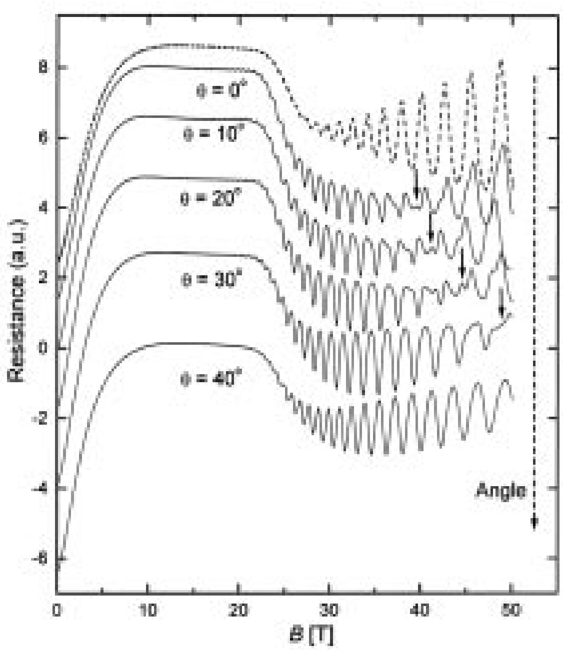

Figure 1. Interplane magnetoresistance for angles θ = 0°, 10°, 20°, 30° and 40° at T = 0.8 K (solid lines) and θ = 0° at T = 3 K (dashed line). The arrows denote the position of the node, indicating the change between *bulk* and *edge* transport mode.

Upon tilting the sample, the node moves up in field, and eventually out of the accessible field range for $\theta > 30^\circ$. This rise of the node position with increasing angle is consistent with the assumption that magnetic breakdown is the mechanism responsible for the destruction of the CFL. Only the effective magnetic field (*i.e.* the component perpendicular to the layers) enters into the argument for the breakdown probability, thereby resulting in a falling probability with increasing angle for a given applied field. Both CFL and QHE, therefore, persist to higher fields for higher angles.

Angle-dependent measurements of the Hall voltage and its sweep rate dependence[6] corroborate this evidence. Pulsed magnetic fields were provided by the NHMFL-Los Alamos. The high-precision rotation insert was developed and assembled in Los Alamos.

References:

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Hall Potential Oscillations in κ-(BEDT-TTF)$_2$I$_3$ in Pulsed Magnetic Fields of Up to 60 T

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Single crystals of the layered, two-dimensional (2D) superconductor κ-(BEDT-TTF)$_2$I$_3$ are among the purest organic conductor samples available. This makes them an ideal test-bed for the investigation of phenomena in bulk crystalline materials of reduced dimensionality. Recent experiments[1] have revealed signatures of the quantum Hall effect (QHE), including saturating eddy current resonances in the magnetization, and a reduction of the amplitude of the Shubnikov-de Haas oscillations in the interplane magnetoresistance at low temperatures. The latter can be interpreted in terms of current-carrying edge states, characteristic of multi-layered quantum Hall systems.[2] Previously, the only other 2D organic conductors to exhibit this type of QHE have been the salts α-(BEDT-TTF)$_2$$M$Hg(SCN)$_4$ (M = K, Tl).[2-4] In these materials, inductive measurements of the Hall potential in pulsed magnetic fields provide essential information about the in-plane resistivity tensor components, and the persistence of the QHE to higher fields.[4, 5]

To measure the inductive Hall potential in κ-(BEDT-TTF)$_2$I$_3$ in magnetic fields of up to 60 T, we have refined the technique applied to α-(BEDT-TTF)$_2$$M$Hg(SCN)$_4$ [4,5] by adding a tunable compensation-coil circuit. In this way, most of the open-loop pick-up was cut off, greatly enhancing the signal-to-noise ratio (Figure 1). The pulsed magnetic fields were provided by the NHMFL-Los Alamos.

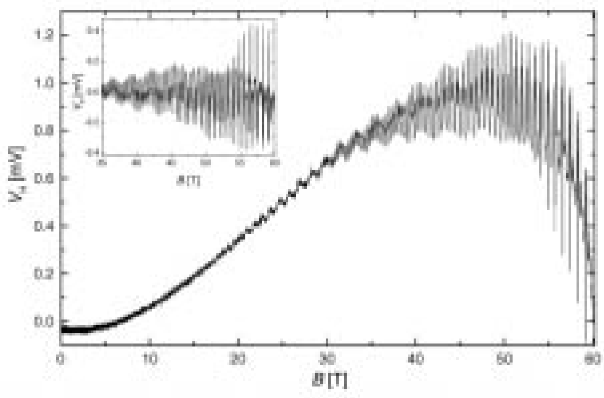

Figure 1. Raw signal of the measured inductive Hall potential V_H at T = 0.5 K, employing an open-loop compensation method. Inset: the oscillatory component of the Hall potential for 35 T < B < 60 T after subtraction of the background.

In Figure 2, we compare the interplane magnetoresistivity ρ_{zz} with the measured Hall potential at fields 48 T < B < 60 T. Since the Hall potential can be expressed as $V_H \approx a^2(\partial B/\partial t)(\rho_{xy}/\rho_{||})$, where a is the sample radius and $\rho_{||} \approx 1/2\,(\rho_{xx} + \rho_{yy})$, V_H is expected to oscillate in phase with ρ_{zz}.[6] This treatment is able to describe the data in the α-phase salts qualitatively.[4, 5] However, some remarkable differences are observed in κ-(BEDT-TTF)$_2$I$_3$:

- The oscillations in ρ_{zz} display a high degree of Zeeman splitting of the Landau levels[7] over the entire range of Figure 2, whereas the Hall voltage oscillations only take on a spin-split appearance for B > 57 T.

- "Spikes" occur in fields at which the chemical potential μ resides within a Landau level.[8] These are especially pronounced for fields coinciding with deep minima in ρ_{zz} (*i.e.* when μ lies within spin-down levels).[9] Smaller peaks also appear for B > 57 T, when μ is in the spin-up levels.

- The size of the "smooth" oscillations in V_H (those oscillating in phase with ρ_{zz}) does not approach the limiting value $\hbar\omega_c$ (≈ 0.9 meV at B ~ 55 T), expected in the QHE regime. The large "spikes" occurring at the minima of V_H, on the other hand, fulfil this condition.

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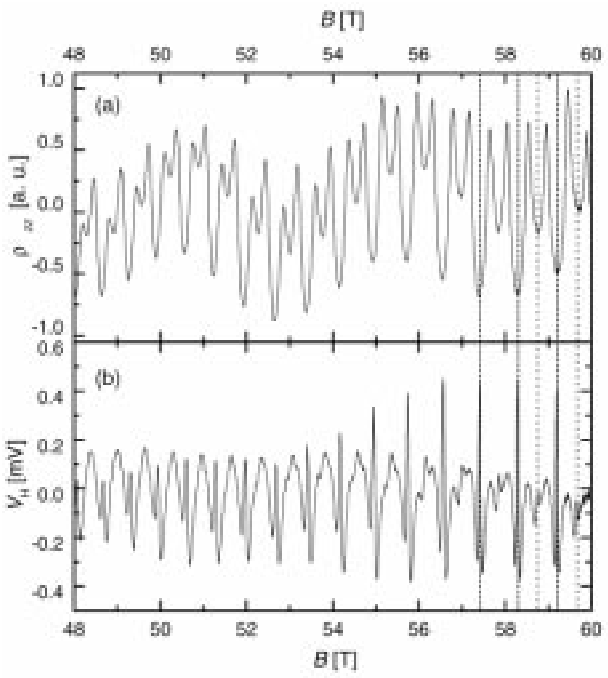

Figure 2. (a) The oscillatory interplane magnetoresistivity for 48 T < B < 60 T; (b) oscillatory part of V_H in the same interval (rising field). The dashed (dotted) lines indicate fields where μ lies within the spin-down (spin-up) Landau levels.

These effects cannot be easily reconciled with previous models.[4] An explanation should be sought by concentrating on the differences between κ-$(BEDT\text{-}TTF)_2I_3$ and α-$(BEDT\text{-}TTF)_2$ $M$$Hg(SCN)_4$ at high magnetic fields, most notably the absence of quasi-one-dimensional states in κ-$(BEDT\text{-}TTF)_2I_3$. This requires a different mechanism for the realization of the QHE.[6]

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9 This can be derived by calculating the filling factor *F/B*, where *F* is the main oscillation frequency.

Magnetic Breakdown in the High Field Phase of the Organic Conductor α-$(BEDT\text{-}TTF)_2KHg(SCN)_4$

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We present new inductive measurements of the differential magnetic susceptibility $\partial M/\partial B$ of the layered organic conductor α-$(BEDT\text{-}TTF)_2KHg(SCN)_4$ in fields of up to 60 T and temperatures down to 400 mK. Clear oscillations of the β-frequency, corresponding to magnetic breakdown (MB) between the quasi-one-dimensional (Q1D) and quasi-two-dimensional (Q2D) sections of the Fermi surface, are seen for the first time at fields above the kink transition.

Pulsed magnetic fields were provided by the NHMFL-Los Alamos. A reduction of the size of the detection system together with a higher attainable peak field enabled a much greater sensitivity to be achieved than in previous studies. To further improve the signal-to-noise ratio, 16 shots were repeated at base temperature and then averaged.

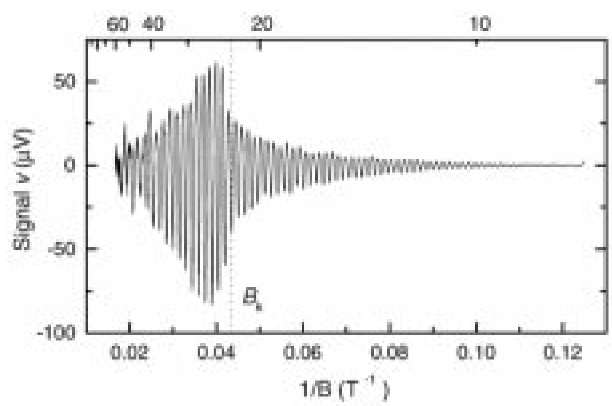

Figure 1. The induced voltage signal at T = 400 mK after averaging several traces. The dotted line indicates the kink transition.

Figure 1 shows the averaged induced voltage $V \propto \partial M/\partial B \cdot \partial B/\partial t$ at T = 400 mK. The kink transition is clearly visible at $B \sim 24$ T. The power spectrum of the oscillations for 24 T < B < 55 T, shows the β-frequency of (4250 ± 5) T and the combination frequencies β-α and β+α (Figure 2), where α is the frequency originating from the Q2D pocket. The application of a semiclassical model, describing the modification of the de Haas-van Alphen amplitude due to MB,[1] enables the MB field B_0 to be determined reliably for the first time, yielding (70 ± 10) T (*c.f.* $B_0 = \sim 265$ T in Ref. 2). This corresponds to an energy gap between the Q1D and Q2D Fermi surface sheets of $\Delta\varepsilon = (23 \pm 2)$ meV.[1, 3] The distance across the gap in k-space can thus be estimated to be $\Delta k_a = 0.27 \cdot 10^9$ m^{-1}. This is only 4 % of the length of the reciprocal lattice vector K_a, as compared to a value of 10 to 20 % predicted by the majority of bandstructure calculations.[4] Magnetotransport measurements of the β-frequency in steady fields above 24 T (Figure 3), carried out at the NHMFL in Tallahassee, yield the same result.[5]

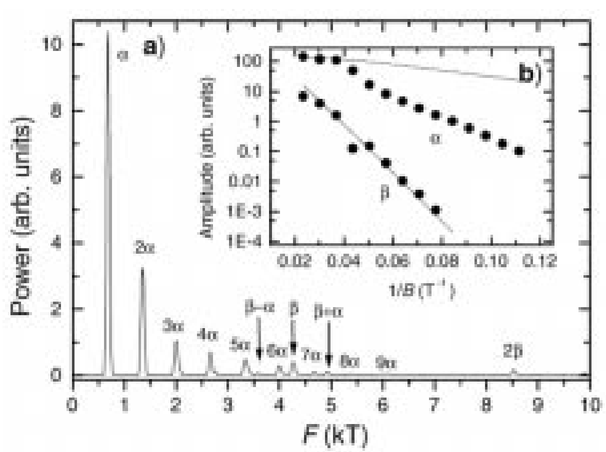

Figure 2. (a) A power spectrum of the oscillations for 24 T < B < 55 T. (b) A Dingle plot of the amplitudes of the α- and β-frequencies. The solid lines show the reconstructed field dependence of the amplitude using the MB field derived in the text. Note that the amplitude of the β-frequency is not attenuated by the transition into the low field state.

In order to accommodate this narrow k-space gap, the Q1D sheets must become more widely separated along the edges of the Brillouin zone. They will, therefore, be more strongly warped than anticipated by bandstructure calculations. An increased warping, however, reduces their ability to nest, and residual pockets within the low-field density wave phase are likely to result from incomplete nesting.[6]

An increased MB probability also has immediate implications for the persistence of the quantum Hall effect to higher fields. With MB becoming more prominent, an increasing number of Q1D carriers will complete the β-orbit, and contribute to the Hall conductivity. The Q1D states, thereby, lose their ability to pin the chemical potential between adjacent Landau levels, leading to a destruction of the quantum Hall effect.[7]

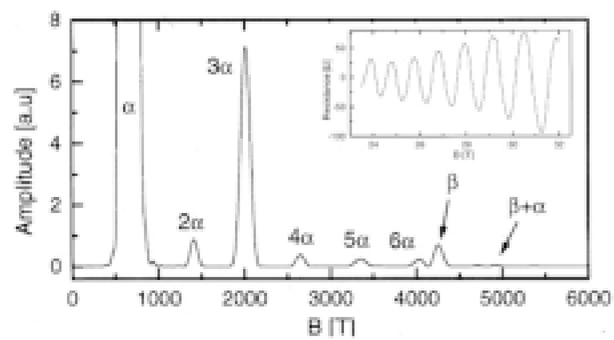

Figure 3. Fourier spectrum of the Shubnikov-de Haas oscillations from steady field transport measurements for 24 T < B < 32 T; the inset shows the trace of the interplane resistance with the background subtracted.

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Pulsed Magnetic Field Measurements of Hall Potential Oscillations in α-(BEDT-TTF)$_2M$Hg(SCN)$_4$ (M = K, Tl) within the Quantum Hall Regime

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We have made direct measurements of the Hall potential in α-(BEDT-TTF)$_2M$Hg(SCN)$_4$ (M = K, Tl) using a variant of the Corbino technique in pulsed magnetic fields of up to 60 T.[1, 2] Rather than applying an external electric field, we use the annular electric field caused by the changing magnetic field of a pulsed magnet, applied normal to the sample's conducting planes; $\partial B/\partial t$ induces a circulating current, which produces a Hall electric field E_H perpendicular to both B and the current. E_H is roughly radial in a cylindrical sample; as long as the charge-transport is ohmic (*i.e.* current $\propto \partial B/\partial t$), $E_H \approx r(\partial B/\partial t)(\rho_{xy}/2\rho_{||})$, where r is the distance from the center and $\rho_{||} \approx 1/2\ (\rho_{xx} + \rho_{yy})$. This leads to a Hall potential $V_H \approx a^2(\partial B/\partial t)(\rho_{xy}/\rho_{||})$ between the center and edge of the sample, where a is the sample radius (Figure 1).

This technique marks the first measurement of its kind. The inductive, wireless provision of the sample current in combination with the high inplane-interplane resistance anisotropy forces the current to flow within the layer. Thus the measured voltage remains free from perturbing interplane contributions, and enables the in-plane components of the conductivity tensor to be determined reliably. These are notoriously difficult to evaluate by other means. This, thereby, circumvents all the main experimental shortcomings of conventional techniques posed by imperfect current contacts, irregular contact and sample geometry, inhomogeneity and defects, in a simple and elegant fashion.

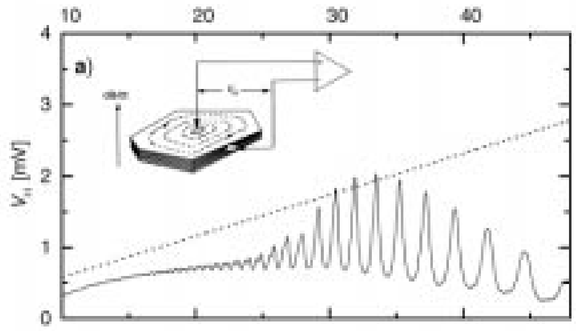

Figure 1. Measured Hall potential in α-(BEDT-TTF)2TlHg(SCN)4 (rising magnetic field; T = 400 mK); similar data are found for the M = K material. The dashed line indicates $\hbar\omega_c$ Inset: sketch of contact arrangement; dashed curves are electric field lines for $\partial B/\partial t$ positive.

Figure 1 shows V_H measured between contacts placed in the center and on the periphery of the sample. Pulsed magnetic fields (~ 10 ms duration) were provided by the NHMFL-Los Alamos. The voltage leads were connected to a high impedance amplifier.

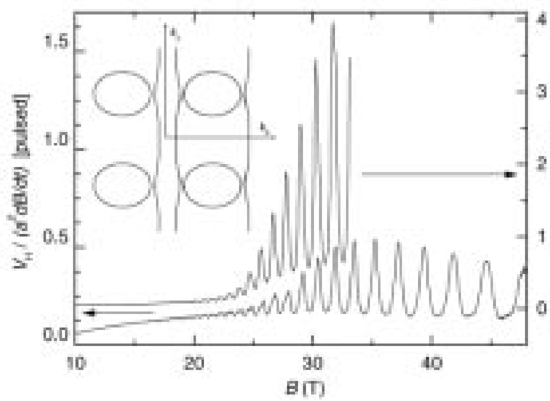

Figure 2. $V_H/a^2(\partial B/\partial t)$ from pulsed field measurements compared with $\rho_{xy}/\rho_{||}$ from steady field data. Inset: schematic of the magnetic breakdown network.

Once the high field phase is reached, strong oscillations occur in V_H. These result from the fact that ρ_{xy} and $\rho_{||}$ oscillate in antiphase, peaks in ρ_{xy} (and thus peaks in V_H) occurring whenever the chemical potential lies between Landau levels.[3, 4] The amplitude of the Hall potential oscillations is roughly of the order of the Landau level spacing $\hbar\omega_c$ (Figure 1). Once the Hall potential energy reaches this limit, the quantum Hall effect (QHE) breaks down due to mechanisms such as Zener tunneling between Landau levels at the sample edges.[5] This leads to a saturation of the induced current at some critical value and, therefore, to a non-linear I-V_{emf} characteristic. Consequently, the in-plane diagonal resistivity $\rho_{||}$ is apparently enhanced within the regime of saturation.

This explains why $V_H/a^2(\partial B/\partial t)$, from pulsed field data, is smaller than $\rho_{xy}/\rho_{||}$ obtained from steady field measurements at the NHMFL-Tallahassee (Figure 2). In the latter case, only the externally applied current is flowing during the measurement (~ 10 μA); this is several orders of magnitude less than the critical current.

The reduction in the size of the oscillations in V_H above 35 T (Figures 1 and 2), on the other hand, is due to the degradation of the QHE caused by magnetic breakdown between the quasi-two-dimensional pocket and quasi-one-dimensional sheets of the Fermi surface[6] (see inset Figure 2). Magnetic breakdown will radically alter the form of the electronic density of states, destroying the arrangement necessary for the realization of the QHE.[4]

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Quantum Oscillations in α-(BEDT-TTF)$_2$KHg(SCN)$_4$ above the Néel Temperature

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Organic charge-transfer salts are model systems for the study of phase transitions characteristic of low-dimensionality. A prime example is the layered conductor α-(BEDT-TTF)$_2$KHg(SCN)$_4$, which forms a density wave (DW) ground state at temperatures below T_N ~ 8 K. At high magnetic fields (B ~ 24 T at T = 0.5 K), the salt undergoes a first-order phase transition manifested by a strong kink in the magnetoresistance, a change in the quantum oscillatory spectrum, and an enhancement of the predominant α-frequency, originating from the quasi-two-dimensional hole cylinder of the Fermi surface.[1]

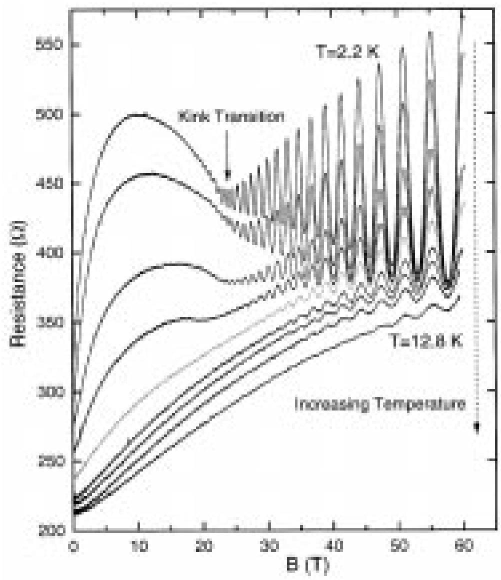

Figure 1. The measured interplane magnetoresistance for temperatures between T = 2.2 K and 12.8 K.

This transition has either been interpreted as a replacement of the DW state by a metallic phase identical with the high temperature-low field state,[1] or as the formation of a separate DW state within a more complicated phase diagram.[2, 3] In order to settle this issue, we have measured the oscillatory component of the interplane magnetoresistivity in pulsed fields of up to 60 T over a wide range of temperatures (0.4 K < T < 13 K),[4] thereby extending the reach of fermiological experiments on this material to far higher temperatures than previously accessible. The pulsed magnetic fields were provided by the NHMFL-Los Alamos.

Figure 1 shows the development of the profile of the magnetoresistance data with increasing temperature. At T = 2.2 K, the pronounced hump in the resistance serves as an indication of the DW state. At higher temperatures, this maximum moves to higher fields and eventually disappears at T ~ 8.2 K. This corresponds to the removal of the DW and is reflected in a change from convexity to concavity of the trace for B < ~ 12 T. The kink transition, marking the phase boundary with the high field state, is blurred out with increasing temperature. As found in previous studies, the complicated low field Shubnikov-de Haas (SdH) spectrum is replaced by a single fundamental frequency (F_α = 667 T) upon leaving the DW phase.

MOLECULAR CONDUCTORS

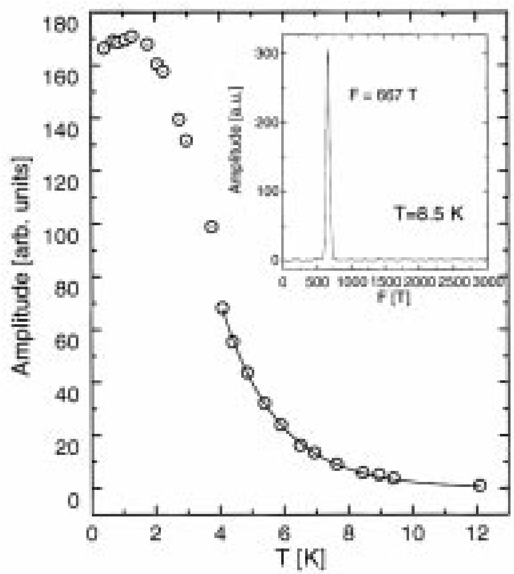

Figure 2. (a) The temperature dependence of the F_α amplitude for 0.4 K < T < 12 K. An LK fit has been applied for T > 4 K. (b) The Fourier spectrum of the SdH oscillations for T = 8.5 K.

The works proposing a non-uniform phase diagram outside the low temperature-low field state predict a further phase transition at high magnetic fields (above 24 T) and temperatures in the vicinity of T_N = 8 K.[2, 3] This area of parameter space had previously been inaccessible. However, no change in the high-field Fourier spectrum is observed as the temperature is raised from 0.4 K up to 12.8 K. The single, dominant peak at F_α = 667 T persists (Figure 2). Similarly, the temperature dependence of the F_α amplitude shows no discontinuity at higher temperatures. A Lifshitz-Kosevich (LK) fit within the temperature range of 4 K < T < 12 K yields an effective mass of $m_\alpha{}^*$ = 1.75(5) m_e, in broad agreement with previous high field studies at low temperatures.[1] Equally, the scattering rate τ^{-1} remains independent of temperature at high magnetic fields.

In summary, we find no evidence for a further phase transition in α-(BEDT-TTF)$_2$KHg(SCN)$_4$ at high magnetic fields with increasing temperature. This corroborates the results of angle-dependent magnetoresistance studies,[1] which showed that the low temperature-high field and the high temperature-low field states are indistinguishable.

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Shubnikov-de Haas and Angle-Dependent Magnetoresistance Oscillation Studies of α-[$(CH_3)_2(C_2H_5)_2N$][Ni(dmit)$_2$]$_2$

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Charge-transfer salts based on the (dmit)-complex[1] are special among two-dimensional organic conductors, since they are thought to be the only ones where the conduction takes place within the planes of acceptor anions.[2] α-[$(CH_3)_2(C_2H_5)_2N$][Ni(dmit)$_2$]$_2$ is the only (dmit)-salt to exhibit quantum oscillations thus far.[3-5] A complete mapping of the Fermi surface (FS) at low temperatures, however, has been thwarted by a series of structural transitions upon cooling,[6] which renders the predictions based on the room-temperature bandstructure inapplicable. Three sets of quantum oscillations have previously been observed, denoted as F_α =10.5 T, F_β = 215 T and F_γ = 4020 T.[3] A fourth series, F_δ = 520 T, has been proposed, but could not be discerned unambiguously from the noise level.[3] In the very high magnetic field limit (30 T < B < 60 T), it has been shown[4] that the Shubnikov-de Haas (SdH) spectrum can be well described by a magnetic breakdown network involving the β- and γ-orbits.

In this report, we present SdH measurements in lower, steady magnetic fields of up to 32 T, provided by the NHMFL-Tallahassee. Figure 1 shows the oscillatory component of the magnetoresistance at T = 0.5 K, together with a Fourier transform performed over the interval 9 T < B < 18 T (see inset). Remarkably, we observe a pronounced peak at F_δ = 520 T, clearly confirming the existence of the corresponding orbit.

In addition to F_β and F_γ (outside the shown window), a new frequency at ~ 31 T can be seen, which we tentatively label F_ε. Although this peak occurs in a region where low-frequency noise owing to the analysis technique is prominent, new measurements down to 100 mK on a different sample appear to confirm the results.[7] In contrast to the β-frequency, neither F_δ nor F_ε can be observed in the high field limit.[4]

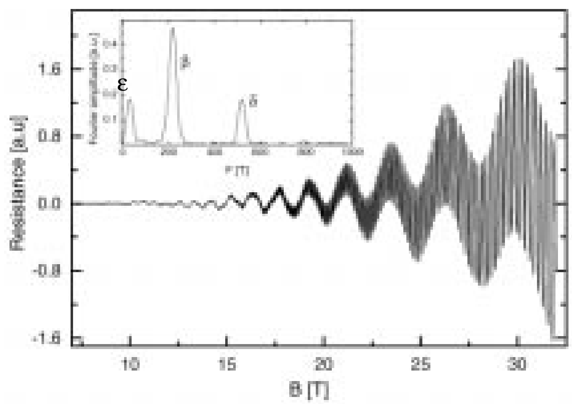

Figure 1. Oscillatory component of the interplane magnetoresistance for T = 0.5 K. Inset: Fourier transform over the field range 9 T < B < 18 T.

A low-temperature FS has been proposed based on crystallographic data taken at 11 K.[6] In this model, a multitude of frequencies are possible, not all of which are observed in our experiments. It is, therefore, essential to map the shape of the main orbits detected experimentally to assess the applicability of the FS model. To this end, we have performed angle-dependent magnetoresistance oscillation (AMRO) studies at a variety of temperatures and magnetic fields. Figure 2 shows a full azimuthal dependence indicating that the features can likely be attributed to a number of quasi-two-dimensional orbits. However, owing to the complexity of the observed structure, analysis is still being undertaken. One of the most striking features is the presence of an enormous spike on each trace at a polar angle of 90°, corresponding to an increase of the magnetoresistance by several orders of magnitude over an angular range smaller than 10 degrees. A comprehensive model is under development,[7] based on a semiclassical Boltzmann approach.[8]

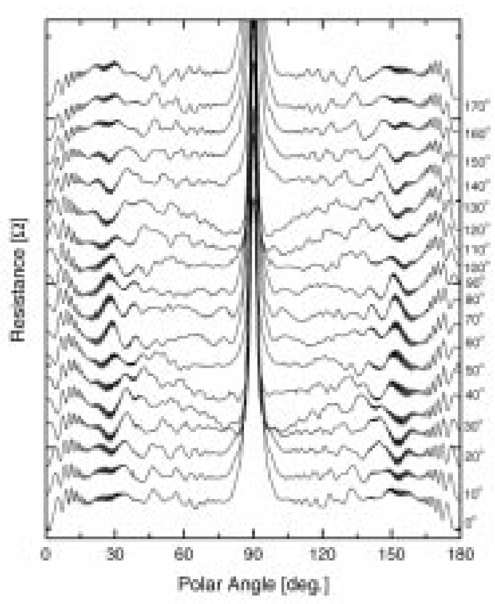

Figure 2. Full azimuthal dependence of the AMRO traces at a constant field of B = 33 T and T = 1.5 K.

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High Magnetic Field ^{13}C NMR in α-(BEDT-TTF)$_2$KHg(SCN)$_4$

NHMFL

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We present NMR measurements on α-(BEDT-TTF)$_2$KHg(SCN)$_4$, in which the six inner carbon sites of BEDT-TTF are labeled with the ^{13}C

isotope, at low temperatures and in magnetic fields up to 28.8 T. We find[1,2] that the density wave ground state of this system persists to fields well above the so called “kink field”, a hysteretic transition observed in transport near 23 T below 6 K (Figure 1). The ^{13}C NMR spectrum is relatively insensitive to crossing the phase boundary, a result that does not support the notion that the ground state is a conventional spin density wave ground state (Figure 2). This result is consistent with previous NMR work on the Rb salt at 8 T.[3] Using the spin relaxation measurements in high magnetic fields (Figure 3), we obtain a BCS-like relationship between the transition temperature and the energy gap, as derived using a simple model (Figure 4). The main implication of this result is that in the low temperature limit, the ground state of this material is not fully metallic, but still partially gapped, even to 28.8 T.

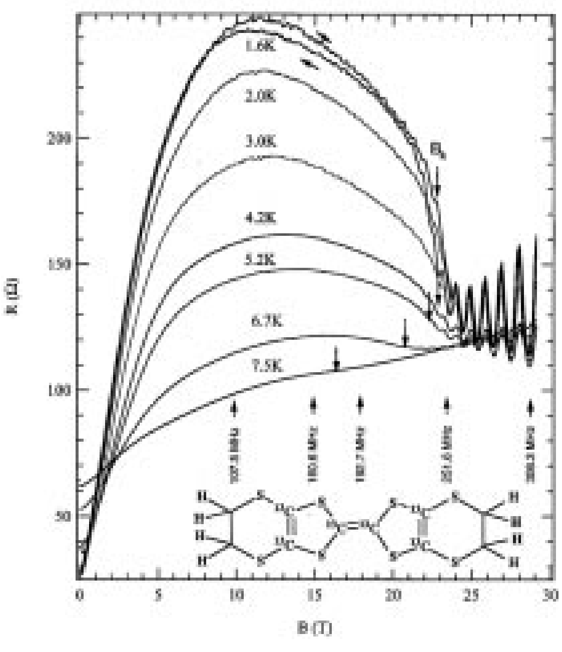

Figure 1. Magnetoresistance of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ vs. magnetic field at low temperatures. Arrows (only shown for 1.6 K data) represent typical hysteretic behavior seen below about 4 K in up and down field sweeps. Down arrows–position of “kink” transition. Up arrows–field values where NMR studies were made. Inset: schematic of BEDT-TTF molecule (planar representation) showing positions of ^{13}C substitutions. These transport measurements were made after the NMR studies were completed (on the same sample).

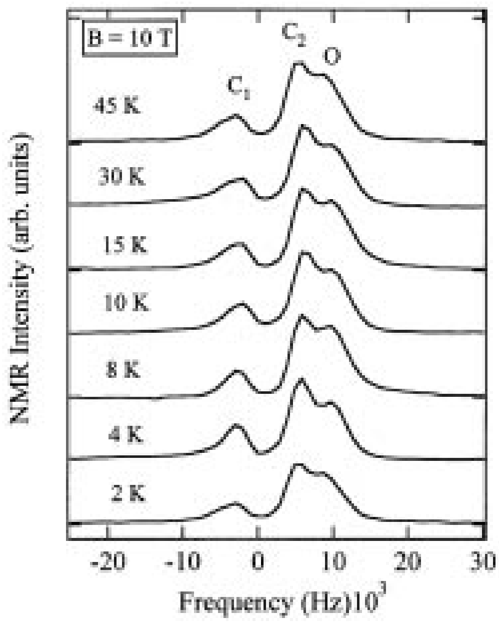

Figure 2. Temperature dependence of the line shape for α-(BEDT-TTF)$_2$KHg(SCN)$_4$ at temperatures above and below T_p = 8 K for the 10 T data. Lines labeled C_1 and C_2 result from the dipolar interaction of the central carbon bond. The line labeled O refers to the out-lying carbon bond, which has negligible dipolar splitting.

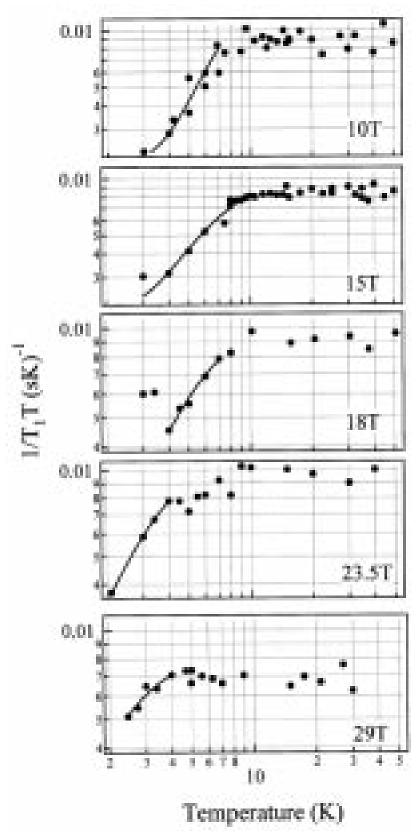

Figure 3. ^{13}C spin lattice relaxation times plotted as $1/T_1T$ vs. temperature (Korringa plot) at each magnetic field studied for α-(BEDT-TTF)$_2$KHg (SCN)$_4$.

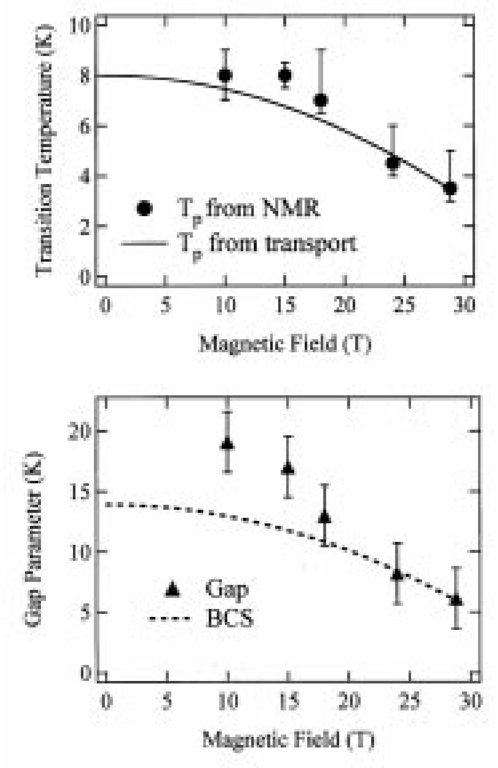

Figure 4. Parameters of α-$(BEDT\text{-}TTF)_2KHg(SCN)_4$.(a) Transition temperature $T_p(B)$ from the present NMR studies compared with the quadratic functional from transport measurements.[1] (b) Gap parameter from fits of Equation 1 to the $1/T_1T$ data of Figure 3. Shown for comparison is the estimated BCS gap based on $T_p(B)$ from transport.

It has been pointed out[4] that in high fields the Landau gap may be manifested in the T_1 data, and this possibility is being considered as the subject for future investigations. In particular, in Figure 1, as the NMR frequency is increased in the field range corresponding to the quantum oscillations, the relaxation rates may also oscillate. Such an effect would be very novel in the area of high field NMR in these organic systems.

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The Superconducting State in $(TMTSF)_2PF_6$ at High Magnetic Field

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From the temperature dependence of the resistivity in the quasi 1D molecular superconductor $(TMTSF)_2PF_6$ at 6 kbar pressure, we showed previously that strong enhancement of the upper critical field, when the field is aligned along the b-axis, leads to a novel crossover between $H_{c2}//a$ and $H_{c2}//b$ near the Pauli pair breaking field (H_{PPB}~2 T).[1] This $H_{c2}//b$ has been reinvestigated at the NHMFL with applied fields up to 17.5 T and with a base temperature of 0.02 K. The pressure was adjusted to be under 6 kbar, but just above a critical pressure for suppression of the spin density wave state ($P_c \approx 5.7$ kbar) where the zero-field resistivity remains metallic ($d\rho/dT>0$) until the superconducting transition. Only one rotation axis was available, so the b*-c* plane was arranged. In a small field along the b-axis, the temperature dependence of the resistivity becomes negative and naturally forms a peak due to the superconducting transition. The peak position at a given field is shown in the Figure 1 as an H-T phase diagram. The data with filled squares ($P=5.7^{+}$ kbar), taken at the NHMFL, are plotted with others for comparison. One of the remarkable features is that the superconductivity persists up to 9 T, about four times the Pauli limiting field. The strong upward curvature with nearly diverging behavior of the critical field at low temperature is independent of the various criteria employed to construct the phase diagram. The ultimate reentrance of super-conductivity has not been found. Due to a partial misalignment due to the single rotation axis, however, even a slight *a*-axis field component can weaken or destroy the field induced dimensional crossover effect. Our observations, strong positive curvature in the upper critical field and survival of the superconductivity well above the Pauli limiting field, are consistent with proposed theories,[2] and strongly suggestive of spin triplet pairing.

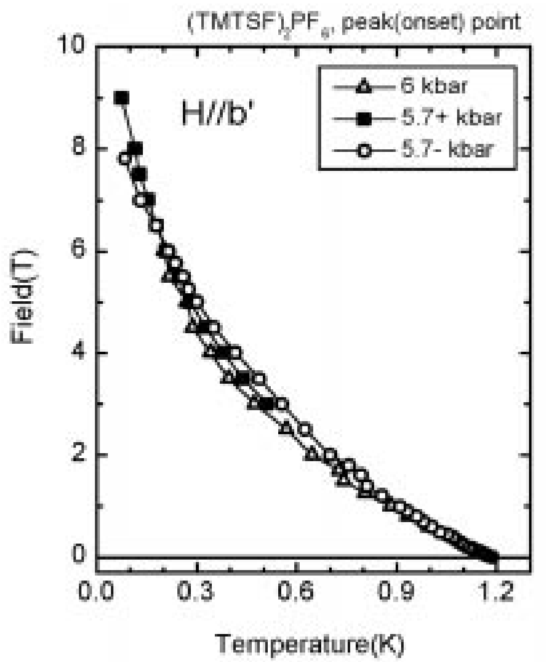

Figure 1. The upper critical phase diagram obtained from the onset (peak) criterion. Strong upward curvature with nearly diverging behavior at low temperature is independent of criterion. Open triangles are for 6 kbar (Ref. 1) and filled squares for slightly above 5.7 kbar, where the normal state resistivity at zero field remains metallic. Open circles are for the pressure slightly below 5.7 kbar, where the zero-field resistivity curve becomes negative below 3.8 K until the superconducting transition at 1.18 K. A complete transition with non-measurable resistance was observed below $\mu_0 H$=6 T within the obtainable temperature range at the pressure P=5.7$^+$ kbar.

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$(TMTSF)_2ClO_4$ in the Quenched State

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In order to preserve the high temperature electronic structure the $(TMTSF)_2ClO_4$ samples are quenched through the anion ordering transition (~ 23 K). Electrical transport contacts were made via silver paint and 12μm gold wire. All of the consecutive temperature measurements (to 50 T) were performed with a single quench to preserve the disorder in the sample.

50 T magnetotransport results are shown in Figure 1. It is clear that quantum oscillations are easily distinguished up to 9.49 K. An interesting kink occurs at high magnetic field that is approximately a monotonic function of field. Although the interpretations of the H-T dependence are preliminary, the kink points appear to be completely uncorrelated to the canonical H-T phase diagram found in $(TMTSF)_2ClO_4$.[2] The

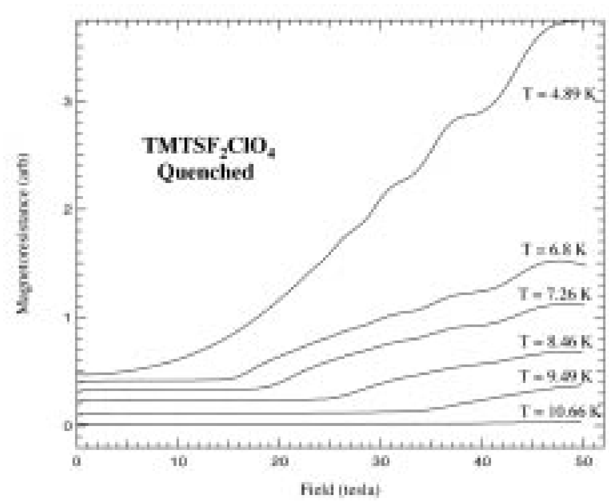

Figure 1. The quenched state magnetotransport of $(TMTSF)_2ClO_4$ measured to 50 T at various temperatures. The quantum oscillations persist to 9.49 K.

quenched state of $(TMTSF)_2ClO_4$ may prove to be as interesting as the annealed state. When $(TMTSF)_2ClO_4$ is quenched through the AO transition, quantum oscillations appear in the high field state and reveal the existence of a high field/high temperature state that has until now been unreported.

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Angle-Dependent Magnetoresistance Oscillation Studies of λ-$(BEDT\text{-}TSF)_2GaCl_4$

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The quasi-two-dimensional (Q2D), layered compound λ-$(BEDT\text{-}TSF)_2GaCl_4$ is the only known superconductor based on the organic donor molecule BEDT-TSF. Derived from their BEDT-TTF[1] sister compounds, BEDT-TSF salts were originally designed to augment the parallels with high-temperature cuprate superconductors by increasing the electronic bandwidth.[2, 3] While most of the synthesized salts, however, remain metallic down to the lowest temperatures, λ-$(BEDT\text{-}TSF)_2GaCl_4$ exhibits a remarkably high superconducting onset temperature of ~ 8 K.[2, 3] It has generated a great deal of interest due to the proposition of multiple-phase superconductivity,[4] and its isostructuralism to BEDT-TSF conductors containing magnetic anions.[5]

Bandstructure calculations for λ-$(BEDT\text{-}TSF)_2GaCl_4$ [3] predict a pair of corrugated, quasi-one-dimensional (Q1D) Fermi surface (FS) sheets and a Q2D hole pocket comprising ~ 33 % of the first Brillouin zone (BZ). Recent transport measurements in pulsed magnetic fields, and at a temperature of T = 370 mK, however, have revealed two sets of Shubnikov-de Haas (SdH) oscillations above 40 T, corresponding to FS pocket areas of ~ 15 % and ~ 95 % of the first BZ.[6] While the latter can be related to a magnetic breakdown orbit between the Q1D and Q2D sections of the FS, the origin of the smaller pocket remains unknown. To this end, we have performed angle-dependent magnetoresistance oscillation (AMRO) studies on λ-$(BEDT\text{-}TSF)_2GaCl_4$. AMRO measurements are uniquely suited to derive *both* area *and* shape of the pocket in question.[7]

Figure 1 shows an azimuthal dependence of the AMRO traces at a constant field of B = 27 T and a temperature of T = 1.5 K. The convexity and peaked shape of the data indicate that the AMROs can be attributed to a Q2D FS pocket .[7] An elliptical callipering fit[8] to the locus of the projection of the Fermi wavevector (Figure 2) yields a pocket of ellipticity $\varepsilon \sim 3$, with an area of $(2.5 \pm 0.2) \cdot 10^{19}$ m^{-2} (~ 60 % of the BZ). This is in disagreement with both the results of the bandstructure calculations[3] and the SdH experiments.[6]

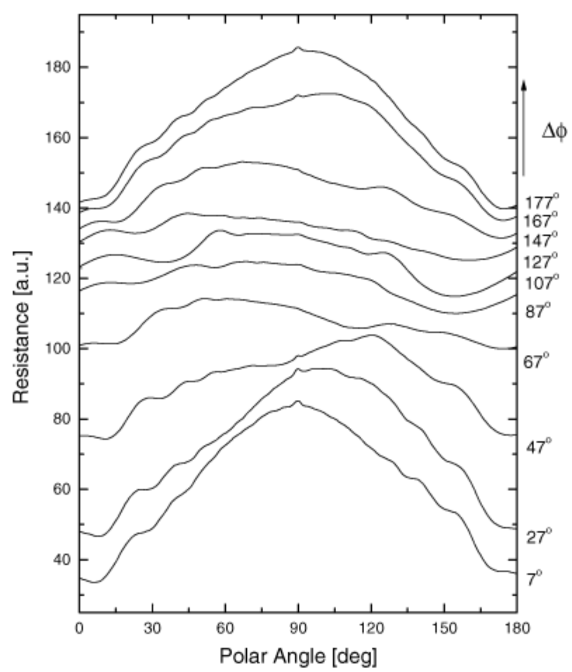

Figure 1. Azimuthal dependence of the AMRO traces at B = 27 T and T = 1.5 K.

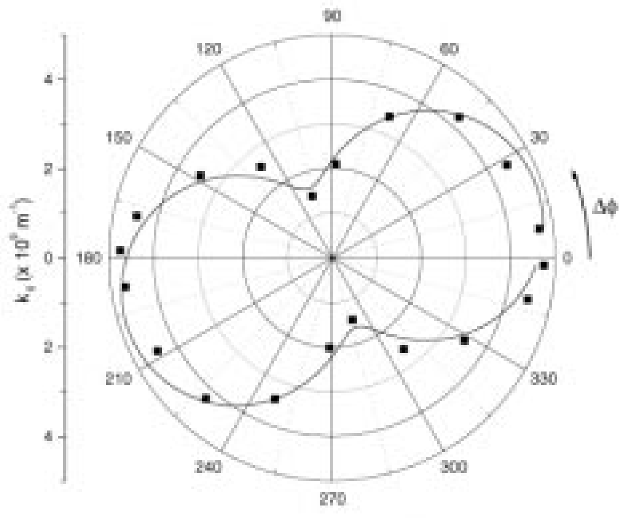

Figure 2. A polar plot of the projection of the Fermi wavevector onto the plane of rotation. The shape of the pocket can be derived from the dumbbell fit by drawing the normal to the radius at each point of the locus.

While the predictions of the band calculations may not be applicable at low temperatures, and high magnetic fields due to thermal contraction of the unit cell,[5] and the possibility of further phase transitions outside the superconducting state, SdH measurements are usually considered to give a reliable image of the FS topology. It is, however, interesting to note that the pocket area extracted from the AMRO fit amounts to four times that derived from SdH oscillations. This allows the conjecture that the difference might be due to a doubling of the interplane lattice constant caused by a folding of the BZ due to a density wave transition. Similar phase diagrams have been found in some TMTSF and BEDT-TTF salts.[9] Clearly more research is needed, however, to support or refute such an assumption. In particular, the advent of higher purity samples should allow a more direct probe of potential phase boundaries via the observation of quantum oscillations at lower fields and higher temperatures.

References:

1 BEDT-TTF stands for bis(ethylenedithio)tetrathiafulvalene; in BEDT-TSF, the affix "thia" is replaced by "selena" to indicate the substitution of selenium as heteroatoms into the inner rings.

2 Kobayashi, H., *et al.*, Chem. Lett., 1993, 1559 (1993).

3 Montgomery, L.K., *et al.*, Physica C, 219, 490 (1994).

4 Mielke, C.H., *et al.*, Phys. Rev. Lett., submitted.

5 Kobayashi, H., *et al.*, Synth. Met., 70, 867 (1995).

6 Mielke, C.H., *et al.*, Phys. Rev. B, submitted.

7 Blundell, S.J., *et al.*, J. Phys. I France, 6, 1837 (1996).

8 For a detailed description of the analytical technique, see A. A. House *et al*., J. Phys.: Condens. Matter, 8, 8829 (1996);

9 Wosnitza, J., *Fermi surfaces of low-dimensional organic metals and superconductors* (Springer, Berlin, 1996).

Magnetotransport Measurements in α-$(BEDT\text{-}TSF)_2FeCl_4$

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We have performed measurements of Shubnikov-de Haas (SdH) and angle-dependent magnetoresistance oscillations (AMRO) in κ-$(BEDT\text{-}TSF)_2FeCl_4$ in fields of up to 33 T provided by the NHMFL-Tallahassee.

κ-$(BEDT\text{-}TSF)_2FeCl_4$ is an organic charge-transfer salt which contains Fe^{3+} magnetic ions and remains metallic at low temperatures.[1] In contrast, its λ-phase sister compound undergoes a metal-insulator transition at 8 K, which is reversed at magnetic fields above 10 T.[2] The electronic structure of the κ and λ phases of $(BEDT\text{-}TSF)_2FeCl_4$ in high magnetic fields has been of growing interest because of the presence of magnetic inorganic layers, and the striking differences between their ground states. The Fermi surface of κ-$(BEDT\text{-}TSF)_2FeCl_4$ has been studied in this report.

Figure 1 shows the SdH oscillations at 450 mK and the inset presents the corresponding frequency spectrum. Fourier transformation of the oscillations reveals two frequencies (F_α= 865 T and F_β = 4230 T) corresponding to a two-dimensional hole pocket, and a complete magnetic breakdown (MB) orbit across the Brillouin zone. Such a frequency spectrum is characteristic of κ-phase salts, and agrees with previous measurements done in the 60 T pulsed magnet at NHMFL-Los Alamos.[3] As shown in Figure 1, however, the classical background magnetoresistance behaves very differently; rather than the negative background resistance,[3] we observed a positive slope to the background. It was suggested that the interaction between conduction electrons and magnetic moments caused the negative magnetoresistance.[3]

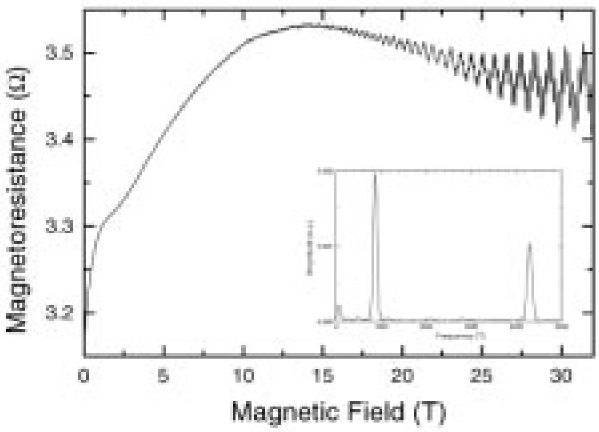

Figure 1. SdH oscillations measured in κ-(BEDT-TSF)$_2$FeCl$_4$ in fields of up to 33 T at 450 mK. Inset: Fourier transformation of the quantum oscillations, yielding the α- and β- frequencies.

Figure 2 presents angle-dependent magnetoresistance oscillations at various relative azimuthal angles Δφ. The traces exhibit a pattern characteristic of a quasi-two-dimensional (Q2D) Fermi surface (FS) with peaks in magnetoresistance corresponding to the large MB β- orbit. Their φ angle-dependence was analyzed,[4] yielding an elliptical Q2D FS pocket with major axis $k_c = (4.12 \pm 0.02) \times 10^9$ m^{-1} and minor axis $k_a = (3.11 \pm 0.03) \times 10^9$ m^{-1}. The area of this pocket is 98.3 % of the first Brillouin zone, corresponding to a quantum oscillation frequency of 4223 T. This is in good agreement with the direct quantum oscillatory measurement of the β- frequency. The exact orientation of the crystal used for this experiment will be determined by infrared-spectroscopy.

In summary, AMRO measurements in κ-(BEDT-TSF)$_2$FeCl$_4$ enable a precise mapping of the magnetic breakdown orbit across the Q1D and Q2D sections of the FS to be carried out.

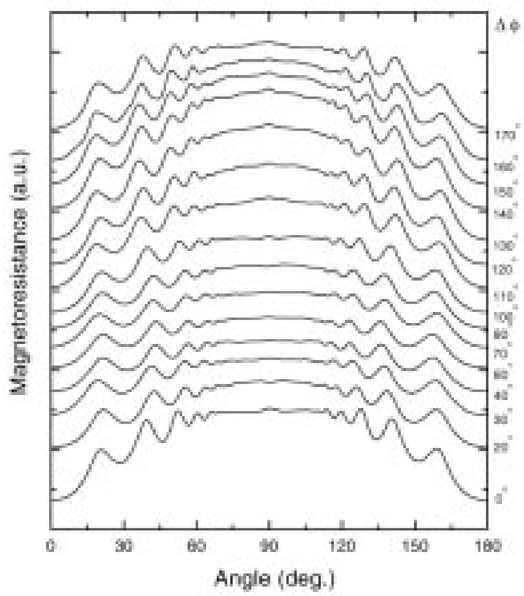

Figure 2. Δφ dependence of the angle-dependent magnetoresistance oscillations in κ-(BEDT-TSF)$_2$FeCl$_4$ at 33 T and 1.6 K.

References:

1 Kobaysahi, A., *et al.*, Chem. Lett., 12, 2179 (1993)
2 Goze, F., *et al.*, Europhys. Lett., 28, 427 (1994).
3 Harrison, N., *et al.*, Phys. Rev. B, 57, 8751 (1998).
4 House, A.A., *et al.*, J. Phys.: Condens. Matter, 8, 8829 (1996)

Observation of the Angle-Dependent Magnetoresistance Oscillation(AMRO) and Shubnikov-de Haas Effect in κ-(BEDT-TSF)$_2$Cu[N(CN)$_2$]Br

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We have carried out angle-dependent magnetoresistance oscillation (AMRO) and Shubnikov-de Haas (SdH) oscillation measurements in κ-(BEDT-TSF)2Cu[N(CN)2]Br in fields of up to 33 T provided by the NHMFL-Tallahassee.

Band structure calculations[1] of κ-(BEDT-TSF)$_2$Cu[N(CN)$_2$]Br predict that its Fermi surface consists of a pair of quasi-one-dimensional (Q1D) Fermi sheets and a quasi-two-dimensional (Q2D) section, the so-called α-pocket. Owing to the relatively small magnetic breakdown (MB) field, B_0 = 10 T, the MB orbit β becomes the dominant electron trajectory at high magnetic fields, establishing a complete breakdown regime.[2] Therefore, no accurate estimation of the size of the pocket using quantum oscillations in high magnetic fields has thus far been possible.

The magnetoresistance of κ-(BEDT-TSF)$_2$Cu[N(CN)$_2$]Br at 450mK is shown in Figure 1. Fourier transforms of the quantum oscillations (presented in the inset of Figure 1) exhibit two frequencies, $F_\alpha = (515 \pm 3)$T and $F_\beta = (3804 \pm 5)$T. The α–frequency represents ~ 13.6% of the Brillouin zone improving earlier estimates from de Haas-van Alphen measurements.[2] It is also in good agreement with the band structure calculations.[1] The β–frequency corresponds to an area equal to that of the first Brillouin zone. Figure 2 shows the azimuthal angle dependence of the AMRO traces. A two-dimensional analysis[3] allows a precise determination of the shape and area of the pocket that gives rise to the AMRO structure. Our analysis yields a Q2D pocket with a minor axis $k_a = 3.29 \times 10^9 m^{-1}$ and a major axis $k_c = 3.40 \times 10^9 m^{-1}$, corresponding to a quantum oscillation frequency of 3690 T. This indicates that the AMRO results from the MB orbit.

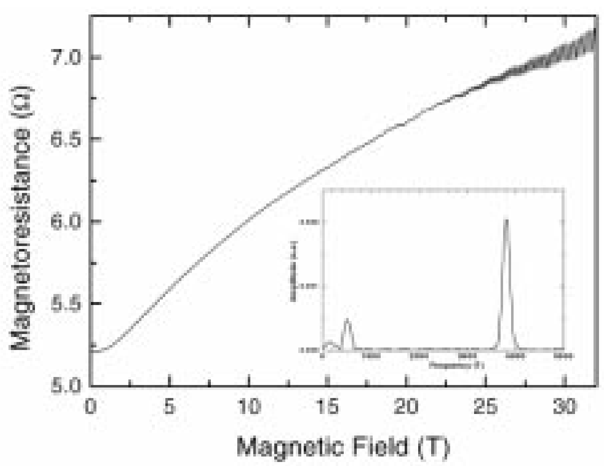

Figure 1. SdH oscillations measured in κ-(BEDT-TSF)$_2$Cu[N(CN)$_2$]Br in fields of up to 33 T at 450 mK. (Inset: Fourier transformation of the quantum oscillations, yielding the α- and β-frequencies.)

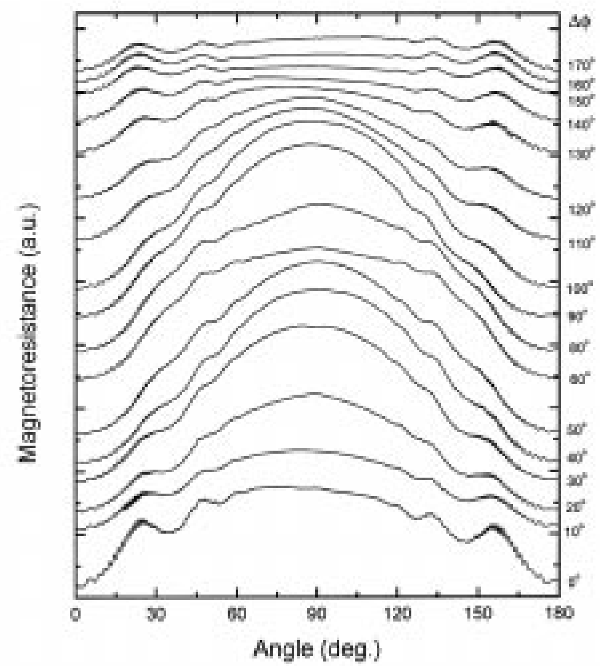

Figure 2. Azimuthal angle dependence of the angle dependent magnetoresistance oscillations in κ-(BEDT-TSF)$_2$Cu[N(CN)$_2$]Br at 33 T and 1.6 K.

References:

1 Burgin, T., *et al.*, J. Mater. Chem., 5, 1659 (1995).
2 Harrison, N., *et al.*, Phys. Rev. B, 56, 12905 (1997).
3 House, A.A., *et al.*, J. Phys.: Condens. Matter, 8, 8829 (1996).

Magnetoresistance of Perchlorate Doped Polyacetylene

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The positive temperature coefficient of resistivity (TCR) from T = 1.5 K to T = 300 K was found in the perchlorate doped polyacetylene (PA).[1] The negative magnetoresistance (MR) was measured for the metallic PA up to H = 17 T. Both transverse ($J \perp H$) and longitudinal (J//H) MR show similar magnetic field dependence, although the magnitude of the transverse MR is larger than that of the longitudinal MR. For the aged sample, the $\rho(T)$ increases more rapidly upon cooling than that of the fresh sample. The MR of the aged sample is positive. The model of interchain charge transfer bridged by the dopants seems to explain the negative MR as well as the metallic temperature dependence of resistivity for the perchlorate doped polyacetylene.

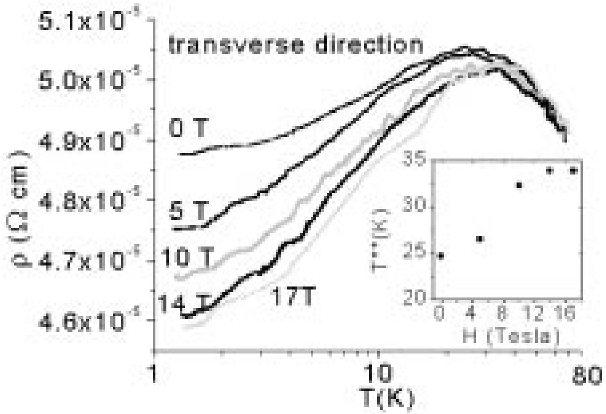

Figure 1. Temperature dependence of the resistivity under fixed magnetic field of the ClO_4^- doped polyacetylene. The inset shows the crossover temperature T** versus magnetic field.

Reference:

1 Park, Y.W., *et al.*, Synth. Met., 96, 81 (1998).

New High Field Angular Dependent Aspects of Quasi-2 Dimensional Organic Conductors

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Organic conductors of α-$(ET)_2MHg(SCN)_4$, where M is an alternating element, show very interesting electronic properties.[1] One approach to understand the behavior in these materials is through Fermi surface (FS) studies.

Angular dependent magnetoresistance measurements (AMRO) performed up to 33 T reveal asymmetric behavior, and large Shubnikov de Haas oscillations (SdH), which display beating effects. The angle θ corresponds to the angle between the field and the b* axis. The angle ϕ corresponds to rotations in the a-c plane. The AMRO behavior can be explained by an asymmetric warping on the Fermi cylinders. The warping can be visualized as rings tilted on a cylinder yet parallel to one another. This model is shown in Figure 2, where D is twice the Fermi vector and A is the warping factor. This kind of symmetry has been suggested before.[2-4] It is easy to see that in this model, AMRO will show asymmetry when rotated parallel to the warping tilt direction, and symmetric behavior when rotated perpendicular to the warping direction.

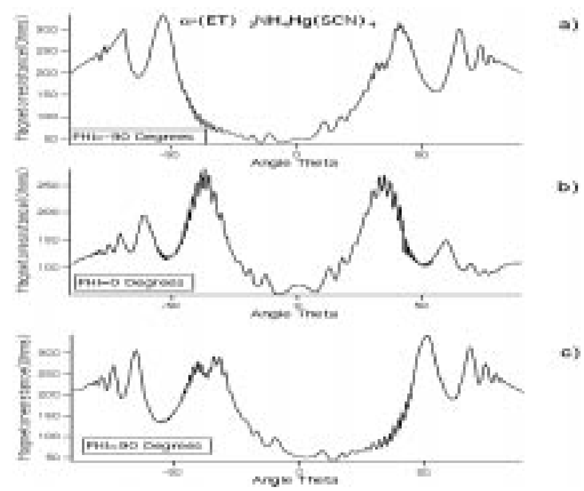

Figure 1. AMRO measurements at 17 T in α-$(ET)_2NH_4Hg(SCN)_4$.

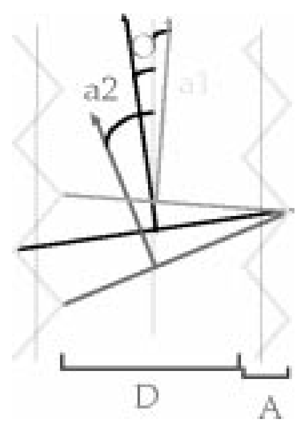

Figure 2. Tilted 2D FS model.

The symmetry about zero θ, in the SdH oscillations, verifies the sample is truly aligned. The offset angle O can be calculated from the angular locations of the first AMRO peaks (a1 and a2) from θ = 0°, by the simple trigonometry relationship:

$$\tan(a2)-\tan(a1) = 2\tan(O)(1+2A/D)/(1+A/D).$$

If we assume D > A, then $\tan(a2)-\tan(a1) \approx 2\tan(O)$. From data, an offset angle of 12.8° is obtained. The central AMRO minimum is actually offset about 13° at $\phi = \pm 90$. Which agrees with the model. Also, the offset angle is close to the 14° angle that exists between the b* and b axis. The M = K material showed additional AMRO structures and behavior, and will be the subject of future studies, as well as, the nature of the warping offset with respect to real crystal structure.

This work was supported by NSF-DMR 95-10427 (JSB).

References:

1 Wosnitza, J., Fermi Surfaces of Low Dimensional Organic Metals and Superconductors, (Springer-Verlag, Berlin, 1996).

2 Yagi, R., *et al*., Solid State Commun., 89, 275 (1994).

3 Hill, S., Phys. Rev. B, 55, 4931 (1997).

4 Kartsovnik, M.V., *et al*., J. Phys. (France) I, 2, 89 (1991).

Temperature and Angular Dependence of the Magnetoresistance in ET_2X Salts

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We have performed systematic measurements of the temperature dependence of the magnetoresistance in α-(BEDT-TTF)$_2$KHg(SCN)$_4$. Strikingly different behavior was observed depending on precise field orientation.

In α-(BEDT-TTF)$_2$KHg(SCN)$_4$, two distinct AMRO regimes have been observed so far. At temperatures above 8 K or applied fields greater than ≈25 T, AMRO features are dominated by behavior identified with a 2D-warped Fermi surface. At lower temperatures and lower fields, the AMRO shows a behavior that is typical of 1D systems. To explore and to better understand both AMRO regimes, we have measured the temperature dependence of single crystals at 14 T and up to 32.5 T at various field positions (see Figure 1).

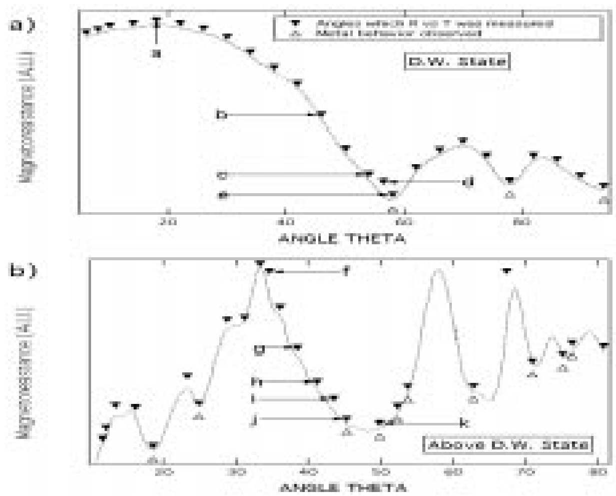

Figure 1. AMRO for α-(BEDT-TTF)$_2$KHg(SCN)$_4$. Down arrows represent points that resistance vs. temperature was measured. (a) AMRO at 14 T, in the density wave state (b) AMRO at 30 T, the high field state.

At AMRO minima, in both regimes, the temperature dependence behavior is metallic to lowest measured temperatures. As the field direction moves away from positions corresponding to an AMRO minima, a nonmetallic cusp begins to appear at low temperature (see Figure 2). This agrees with previously reported data.[1-3] This cusp continues to grow in magnitude and slope as the field direction is changed toward the AMRO maxima. Similar effects have been observed in quasi-one dimensional organic system.[4]

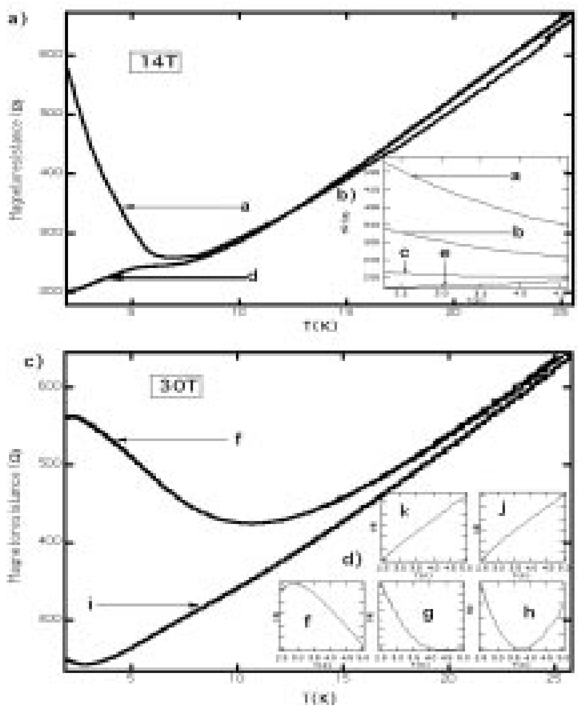

Figure 2. Temperature dependence of magnetoresistance at various field positions from Figure 1.

At high fields, when the measurement is made exactly on AMRO maxima, a new metallic dip forms at low temperatures. Similar temperature dependent effects have been seen by Kartsovnik.[5]

This work was supported by NSF-DMR 95-10427 (JSB). It is clear that future work needs to be continued on this.

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3. Biskup, N., *et al.*, Solid State Commun., 107, 503 (1998).
4. Chashechkina, E.I., *et al*., Phys. Rev. Lett., 80, 2181 (1998).
5. Kartsovnik, M.K., *et al.*, Synth. Metals, 86, 1933 (1997).

Temporal Processes in Complex-Anion Organic Superconductors

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We have systematically studied the time dependent relaxation of the resistivity of κ-(BEDT-TTF)$_2$Cu[N(NC)$_2$]Br, the highest ambient pressure transition temperature (T_c = 11.6 K), and the organic superconductor in its metastable regime between 60 K and 85 K. We identified the relevant ordering parameter from relaxation to the stable state in terms of the amplitude of the time dependent resistivity. This ordering parameter is directly correlated to the superconducting transition temperature, and also to the amplitude of the quantum oscillations associated with the Shubnikov-de Haas effect. This finding sheds new light on mechanisms for the scattering times in this material. We describe a two-level model, which provides an excellent account for the observed temperature dependence of the relaxation process.

After appropriate ordering as depicted in Figure 1, the quantum oscillations associated with the breakdown orbit, although small against the background resistance, are clearly visible at fields as low as 25 T (Figure 2). The insets demonstrate the systematic increase in SdH amplitude with temperature, as expected.

The κ-(BEDT-TTF)$_2$Cu[N(NC)$_2$]Br samples we have investigated exhibit superconductivity and SdH oscillations. Both properties depend on an ordering parameter that is correlated to time dependent changes in the resistivity as observed in a narrow range of temperature (Figure 1). In comparative studies of the relaxation process in zero and in high (30 T) magnetic fields, and also in SQUID magnetometer studies, no evidence of magnetic effects or signatures were observed. We conclude that, for these samples, even in the presence of impurities where the mean free path (from the Dingle temperature)[1] is < 100 nm, the mechanism for relaxation is structural in nature, and involves two different molecular configurations separated by an energy of order 0.2 eV. This energy is of the order of the bandwidths computed in tight binding calculations.[2] Although deuterated samples give some indication that the ethylene groups may be involved in the ordering, the exact assignment is still an open question. Our observation of two relaxation time constants may indicate the participation of multiple molecular sites. Other aspects of the behavior of this material, which remain elusive, are the attenuation of the closed orbit frequency in the SdH amplitudes, and the relatively small SdH amplitude for the breakdown orbit as compared to the magnetoresistance.

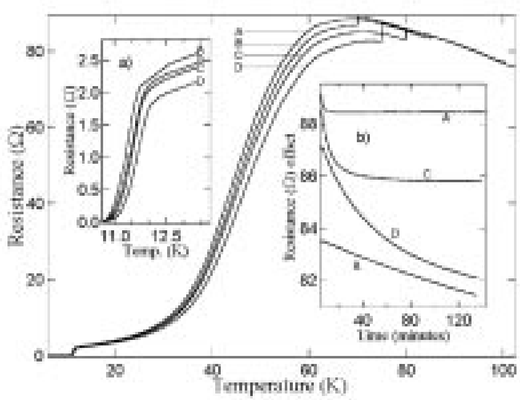

Figure 1. Resistive ordering: κ-(BEDT-TTF)$_2$Cu[N(NC)$_2$]Br resistance vs. temperature with 140 minute holds, varying T_h: A-85 K; B-70 K; C-80 K; D-75 K. (a) Expanded view of superconducting transitions. (b) Resistance vs. time at various T_h's showing exponential behavior.

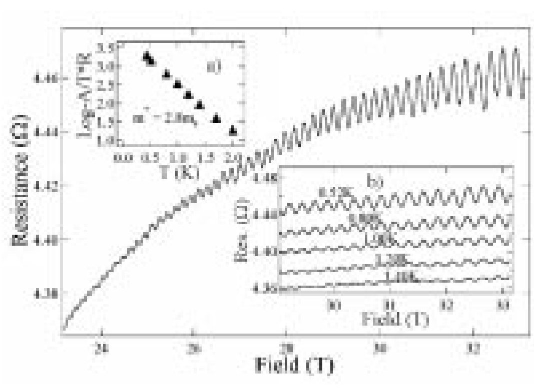

Figure 2. Shubnikov-de Haas oscillations at high fields and 0.45 K. (a) Lifshitz-Kosevich analysis. (b) SdH oscillations at various temperatures.

References:

1 Mielke, C.H., *et al.*, Phys. Rev. B, 56, R4309 (1997).
2 Ching, W.Y., *et al.*, Phys. Rev. B, 55, 2780 (1997).

Upper Critical Field and High Field Magnetoresistance of $Tl_2Mo_6Se_6$

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Kebede, A., North Carolina A&T Univ., Physics
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We have investigated the temperature (T) dependence of the upper critical field of $Tl_2Mo_6Se_6$ (20 mK to 7 K) in magnetic fields up to 60 T. The study was conducted using the 20 T dc magnets at Los Alamos and Tallahassee, and the short pulse magnets at Los Alamos. The results are summarized in Figure 1. The T dependence of the transverse critical field confirms Brusetti's results[1] and has an unconventional behavior with H_{c2} showing a positive curvture at low T, H_{c2}(20 mK) = 5 T. As reported previously, however, in the longitudinal direction, H_{c2} behaves in a conventional manner, with $H_{c2}(0)$ = 23 T, and no upturn is observed down to 20 mK. In the normal state, above H_{c2}, R(H) decreases linearly with increasing field, up to the highest field explored (60 T). In this regime, the slope dR/dH is non-linear with I and decreases when I increases. The two most interesting mechanisms that can lead to this negative slope are (1) H induced restoration of superconductivity at high magnetic fields, and (2) the H-induced suppression of a density wave state. Indeed, previous study suggests that the ground state of this compound is characterized by the competition and coexistence of superconductivity and DW.[1,2] Preliminary study of the IV characteristics in this regime show the conductivity increases when I increases. This suggests the most likely mechanism is the density wave state. Further work is in progress to clarify this issue. Future research will also include the study of the ground state in this compound using uniaxial stress.

This work was supported by a grant from the NHMFL and the NSF, Grant Number DMR9312530.

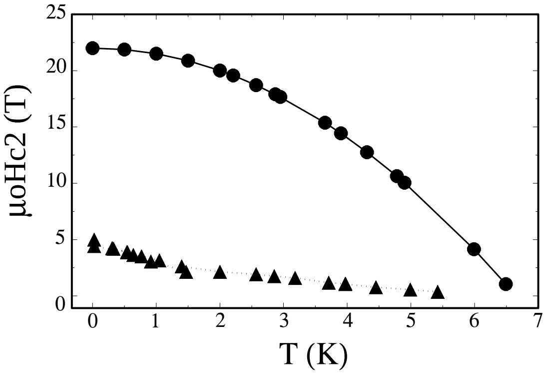

Figure 1. Temperature dependence of the upper critical field. The full triangle and the full circles correspond to the longitudinal and transverse critical field, respectively.

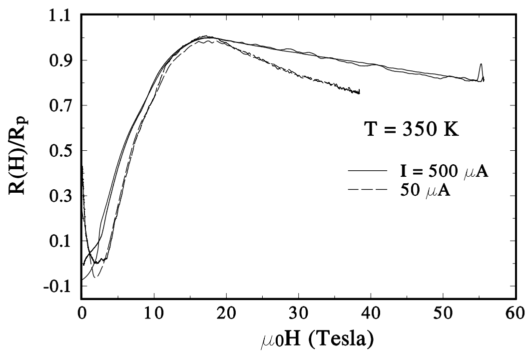

Figure 2. R vs. B at 350 mK. The experiment was conducted using a lock in technique at 517 kHz. Note the current dependence of the negative dR/dH.

References:

1 Brusetti, R., *et al.*, Phys. Rev. B, 49, 8931 (1994-I).
2 Tessema, G.X., *et al.*, Phys. Rev. B, 43, 3434 (1991).

Magnetic Quantum Oscillations in Protonated and Deuterated κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br

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The quasi-two-dimensional metal κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br is the organic superconductor with the to date highest transition temperature of $T_c \approx 11.5$ K. Some recent experiments have revealed unusual superconducting properties and unconventional behavior was suggested.[1] Until very recently no experimental information on the electronic band structure was available. During the last year two experiments on ambient-pressure Shubnikov-de Haas (SdH) oscillations were reported.[2,3] Both experiments give only information for magnetic fields applied perpendicular to the conducting planes and differ somewhat in the experimentally determined effective cyclotron mass (5.4 m_e [3] and 6.4 m_e [2]). In recent experiments up to 30 T at the NHMFL in Tallahassee we were able to detect SdH oscillations in a protonated and (for the first time) in a deuterated sample of κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br for an angular range of up to about 20°. Figure 1 shows the measured resistivities at about 0° and in the insets the clearly visible SdH signals in the relative conductivity Δσ/σ. The studied samples (especially the protonated

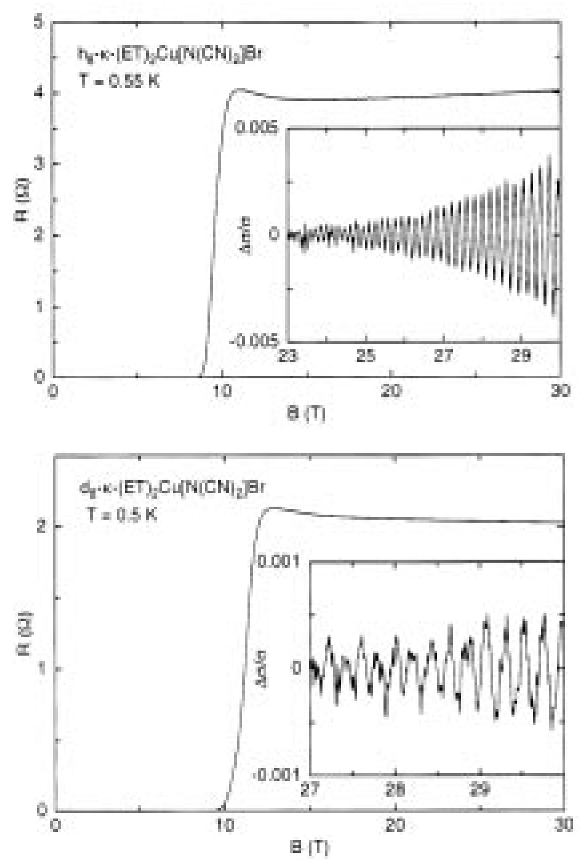

Figure 1. Resistivity and SdH signal of the protonated (top) and deuterated (bottom) sample.

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sample with a residual resistivity ratio of about 100) are characterized by their high quality. The obtained SdH frequencies are in line with the reported values (for protonated samples)[2,3] and follow the expected 1/cosΘ behavior for two-dimensional metals. The effective masses were about 6 m_e for the deuterated sample and 7.3 m_e for the protonated one.

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Unusual Shubnikov–de Haas Oscillations in the Organic Superconductor β"-$(ET)_2SF_5CH_2CF_2SO_3$

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Recently β"-$(ET)_2SF_5CH_2CF_2SO_3$, a new organic superconductor with T_c = 4.5 K (bulk value, diamagnetic onset ~5.2 K) based on the donor molecule bis(ethylenedithio)-tetrathiafulvalene (BEDT-TTF or ET for short) and a purely organic discrete anion, was synthesized.[1] We recently were able to detect the Shubnikov-de Haas (SdH) and de Haas-van Alphen (dHvA) effect in this quasi-two-dimensional (2D) organic superconductor.[2,3] Both kind of experiments (dHvA data up to 26.6 T and SdH data up to 13 T) were in good agreement with predictions from the three-dimensional (3D) Lifshitz–Kosevich (LK) formula and gave oscillation frequencies of F_0 = (199±2) T/cosΘ and effective cyclotron masses of m_{c0} = (1.90±0.05)m_e/cosΘ, where Θ is the angle between field and the normal to the ET planes. Very recent SdH experiments up to 33 T at the NHMFL in Tallahassee, however, revealed highly unusual behavior (see Figure 1). At low temperatures the longitudinal background magnetoresistance (current parallel to the field) shows at Θ = 0 an enormous increase (a factor of ~25 at T = 0.5 K) without a tendency of saturation. Further, the temperature dependence of the SdH amplitude, which is given by the oscillatory part of the conductivity Δσ/σ, deviates dramatically from the usual 3D LK behavior (inset of Figure 1). At high fields above about 20 T the oscillation amplitude increases first with T before the usual SdH amplitude reduction is observed at higher T. Both effects are presumably related to the two dimensionality of the electronic structure and the closeness to the quantum limit. At 30 T only the six lowest Landau levels are occupied. A similar SdH-amplitude damping was observed in κ-$(ET)_2I_3$.[4] The origin of this unusual effect is unclear and further experiments are necessary to elucidate the electronic properties of quasi-2D metals at high fields.

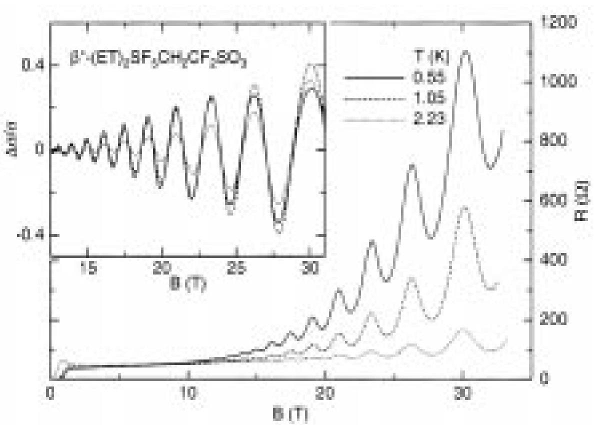

Figure 1. Resistivity and SdH signal up to 33 T. The SdH amplitude decreases with decreasing T.

References:

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